

Monte-Carlo Simulation Studies on the Superspin
Structure of 3D Nanoparticle Supercrystals

Mauricio Cattaneo

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Prof. Dr. David DiVincenzo
Prof. Dr. Thomas Brückel

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Abstract

Nanoparticle (NP) supercrystals constitute a fascinating novel type of material with tunable magnetic, electronic and optical properties [3, 8, 12]. By choosing different NP materials, e.g. ferromagnetic or antiferromagnetic, a variety of magnetic and eventually multifunctional properties might be achieved. Hereby, one major challenge is the deliberate control of the supercrystal structure and of the resulting physical properties. In simulations we are able to model the collective magnetic ground states from microscopic assumptions [3–5]. This thesis aims to develop a proper simulation methodology to deal with lattices of interacting magnetic NP moments. Such a study is a crucial step towards predicting magnetic ground states and energy landscapes as function of the supercrystal lattice type and as function of the individual NP properties. Immediate goals include the study of the influence of dipole-dipole interactions on superparamagnetism and the spin structure of supercrystals at low temperatures.

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1 Introduction

NP supercrystals are regular arrangements of NPs in complete analogy to crystals in condensed matter. An example is given in figure 1.

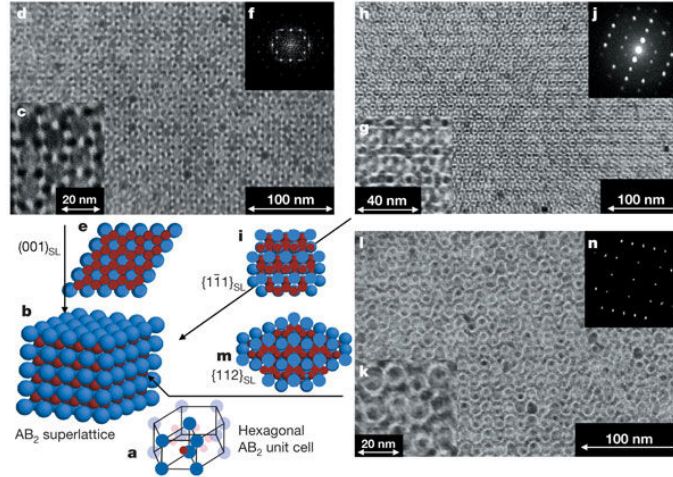


Figure 1: TEM micrographs and sketches of AB_2 superlattices (isostructural with intermetallic phase AlB_2 , SG 191) of 11-nm $\gamma\text{-Fe}_2\text{O}_3$ and 6-nm PbSe NCs. [17]

The research on supercrystals started already several decades ago in the context of colloidal crystals on micron sized silica or polystyrene spheres [11]. In recent years, groups have succeeded in fabricating one, two or three dimensional arrangements of e.g. magnetic or semiconducting NPs using various self-organization techniques [3, 4, 8, 12]. The properties of these systems are determined both by the individual NPs and the interactions among them. Provided the composition and interaction of different NPs can be tuned, interesting behavior and novel applications may emerge [18].

But also from a fundamental point of view, such supercrystals of interacting magnetic nanoparticles are a fascinating subject. In particular NPs which consist of one magnetic single-domain per NP represent an interesting model system. The question whether systems with long-range interaction such as dipole-dipole interactions can exhibit long-range ordered ground states is not comprehensively answered in more than two dimensions. In fact, even the Mermin-Wagner theorem, answering the question for systems of up to two dimensions with a general ‘no’, has been challenged by e.g. Kosterlitz-Thouless transitions, via a divergence of the correlation length [9]. Albeit NP supercrystals have much larger length scales than usually considered in many-body theory, mesoscopic systems do provide very similar questions. The different length-scale and the dominance of the dipole-dipole interaction between the NP macromoments potentially leads to new possibilities of magnetic ordering within physical systems. In any case, the NP magnetic dipole-moments are subject to frustrated interactions.

2 Theory of Magnetism

Theoretical expectations of magnetic measurements will be the cornerstone of this chapter.

One can classify the magnetic phenomena into three main groups:

- Diamagnetism
- Paramagnetism
 - Localized Moments
 - Itinerant Moments
- Collective Magnetism
 - Ferromagnetism
 - Ferrimagnetism
 - Antiferromagnetism

2.1 Origin of Magnetism in Solid State Matter

2.1.1 Bohr-van Leeuwen theorem

Often, para- and diamagnetism are explained as an induction effect. It implies a theory of moving charges which can be treated within a classical atom model, and vector analysis as the significant mathematical language to understand magnetism. The general idea might be the following: The Larmor precession of the orbital angular momentum around the direction of the magnetic field induces an extra moment which according to Lenz's law is directed oppositely to the orientation of the applied field. Trying to rigorously calculate any atomic magnetic moment in this setting will inevitably lead to a contradiction as we will see now:

Let a solid consist of identical ions and let it possess translational symmetry. We can then write the magnetization as

$$\mathbf{M} = \frac{N}{V} \langle \mathbf{m} \rangle$$

where $\mathbf{m}$ is the magnetic moment of the individual ion. N is the number of ions in the volume V . If magnetism is a classical phenomenon, then each ion in our solid must offer a classical Hamiltonian function H and we have the following relations from statistical mechanics:

$$\mathbf{m} = -\nabla_{\mathbf{B}_0} H \quad (2.1)$$

$$\langle \mathbf{m} \rangle = \frac{1}{Z} \int d^{3N_e} x \int d^{3N_e} p \mathbf{m} e^{-\beta H} \quad (2.2)$$

$$Z = \frac{1}{N_e! h^{3N_e}} \int d^{3N_e} x \int d^{3N_e} p e^{-\beta H} \quad (2.3)$$

where

- Z is the classical partition function
- N_e is the number of electrons per ion
- $\beta \equiv (k_B T)^{-1}$ is the inverse temperature
- $\frac{1}{N_e! h^{3N_e}} \int d^{3N_e} x \int d^{3N_e} p \equiv \int_{\Gamma} d\gamma$ is the normalized integration over the complete phase space spanned by N_e electrons (3 dimensions for both position and momentum variables).

We arrive at

$$\begin{aligned} \langle \mathbf{m} \rangle &\stackrel{(2.2)}{=} \frac{1}{Z} \int_{\Gamma} d\gamma e^{-\beta H} \mathbf{m} \stackrel{(2.1)}{=} -\frac{1}{Z} \int_{\Gamma} d\gamma \underbrace{e^{-\beta H} \nabla_{\mathbf{B}_0} H}_{= -\frac{1}{\beta} \nabla_{\mathbf{B}_0} e^{-\beta H}} \\ &= \frac{1}{\beta Z} \nabla_{\mathbf{B}_0} \int_{\Gamma} d\gamma e^{-\beta H} \stackrel{(2.3)}{=} \frac{1}{\beta Z} \nabla_{\mathbf{B}_0} Z \end{aligned} \quad (2.4)$$

We therefore need to investigate the field dependence of the classical partition function. Noting that any magnetic field $\mathbf{B}_0$ can be written as $\mathbf{B}_0 = \nabla \times \mathbf{A}$, we rewrite our Hamilton function as

$$H = \frac{1}{2m} \sum_{\alpha=1}^3 \sum_{i=1}^{N_e} ((\mathbf{p}_i)_{\alpha} + e \mathbf{A}_{\alpha})^2 + H_{\text{int}}(\mathbf{r}_1, \dots, \mathbf{r}_{N_e})$$

where H_{int} is the term representing the electron interactions. Thereby, we separated the position- and momentum dependent parts of the total Hamilton function which

lets the partition function take the form

$$Z = \frac{1}{N_e! h^{3N_e}} \int_{\Gamma_{\mathbf{r}}} d^{3N_e} x e^{-\beta H_{\text{int}}(\mathbf{r}_1, \dots, \mathbf{r}_{N_e})} \times \\ \times \int_{\Gamma_{\mathbf{p}}} d^{3N_e} p \exp \left(-\frac{\beta}{2m} \sum_{\alpha=1}^3 \sum_{i=1}^{N_e} ((\mathbf{p}_i)_{\alpha} + e\mathbf{A}_{\alpha})^2 \right)$$

Since the $\Gamma_{\mathbf{p}}$ integration lets every momentum coordinate run from $-\infty$ to ∞ , the canonical momentum

$$\tilde{\mathbf{p}}_i := \mathbf{p}_i + e\mathbf{A}$$

can be transformed linearly with arbitrary but constant $\mathbf{A}$, without changing the integration limits. Since any applied magnetic field $\mathbf{B}_0$ would hence not alter the $\mathbf{p}$ -integration, we have

$$\nabla_{\mathbf{A}} Z = \nabla_{\mathbf{B}_0} Z = 0 \quad (2.5)$$

$$\stackrel{(2.4)}{\Rightarrow} \langle \mathbf{m} \rangle = 0 \quad (2.6)$$

for any magnetic field.

We have therefore shown rigorously that, classically, there is no magnetism.

There cannot be any discussion whether magnetism does exist, therefore we have shown that it must be an effect only understandable quantum mechanically. This is the famous Bohr-van Leeuwen theorem:

Bohr-van Leeuwen Theorem

Magnetism is a quantum mechanical effect. Strictly classically, there cannot be either dia-, para- or collective magnetism

For the purposes of this thesis however, this does not mean that we will proceed to argue strictly quantum mechanically.

We can continue using classical or semiclassical models and calculations.

2.2 Magnetic Interactions

Different types of magnetic interactions are discussed in this section. We focus on interactions that are present in systems composed of magnetic moments of constant length.

2.2.1 Dipole-Dipole Interaction

The first interaction which might be expected to play a role is the magnetic dipolar interaction. Two magnetic dipoles $\mathbf{m}_1$ and $\mathbf{m}_2$ separated by $\mathbf{r}$ have an energy equal to

$$\begin{aligned} E_{\text{dip}} &= \frac{\mu_0}{4\pi r^3} \left[\mathbf{m}_1 \cdot \mathbf{m}_2 - \frac{3}{r^2} (\mathbf{m}_1 \cdot \mathbf{r})(\mathbf{m}_2 \cdot \mathbf{r}) \right] \\ &= \frac{\mu_0}{4\pi r^3} [\mathbf{m}_1 \cdot \mathbf{m}_2 - 3(\mathbf{m}_1 \cdot \hat{\mathbf{r}})(\mathbf{m}_2 \cdot \hat{\mathbf{r}})] \\ \text{with } \hat{\mathbf{r}} &\equiv \frac{\mathbf{r}}{r} \end{aligned}$$

which therefore depends on their separation and their degree of mutual alignment. If the magnetic moments in question are single electrons and we take distances at atomic length scales, the energies were equivalent to roughly 1K in temperature. Therefore, properties of condensed matter are conventionally not overly dependent on the dipole-dipole interaction except for those ordering at mK temperatures. On the other hand, this is a long-range interaction where the complete sample needs to be taken into account. For example, it is conceivable that the specific length scale of this interaction influences the emergence of magnetic domains or other phenomena of magnetic ordering.

2.2.2 Exchange Interaction

In atomic crystals or any conventional magnetic system, exchange interactions are usually responsible for long-range magnetic order. They are purely quantum mechanical in nature, but the underlying principle is electrostatics.

Consider a simple model with just two electrons which have spatial coordinates $\mathbf{r}_1$ and $\mathbf{r}_2$ respectively. The wave function for the joint state can be written as a product of single electron states, so that if the first electron is in state $\psi_a(\mathbf{r}_1)$ and the second electron is in state $\psi_b(\mathbf{r}_2)$, then the joint wave function is in $\psi_a(\mathbf{r}_1)\psi_b(\mathbf{r}_2)$.

Since electrons are fermions, the true wave function must be antisymmetric with respect to particle exchange. So the *spin part* of the wave function must either be an antisymmetric singlet state χ_S ($S = 0$) in the case of a symmetric spatial state or a symmetric triplet state χ_T ($S = 1$) in the case of an antisymmetric spatial state. Therefore we can write the wave function for the singlet case Ψ_S and the triplet case Ψ_T as

$$\begin{aligned} \Psi_S &= \frac{1}{\sqrt{2}} [\psi_a(\mathbf{r}_1)\psi_b(\mathbf{r}_2) + \psi_a(\mathbf{r}_2)\psi_b(\mathbf{r}_1)] \chi_S \\ \Psi_T &= \frac{1}{\sqrt{2}} [\psi_a(\mathbf{r}_1)\psi_b(\mathbf{r}_2) - \psi_a(\mathbf{r}_2)\psi_b(\mathbf{r}_1)] \chi_T \end{aligned}$$

where both the spatial and spin parts of the wave function are included. If we assume the spin parts to be normalized, the energies of the two possible states are

$$\begin{aligned} E_S &= \int \Psi_S^* \mathcal{H} \Psi_S d^3r_1 d^3r_2 \\ E_T &= \int \Psi_T^* \mathcal{H} \Psi_T d^3r_1 d^3r_2 \\ \Rightarrow E_S - E_T &= 2 \int \psi_a^*(\mathbf{r}_1) \psi_b^*(\mathbf{r}_2) \mathcal{H} \psi_a(\mathbf{r}_2) \psi_b(\mathbf{r}_1) \end{aligned} \quad (2.7)$$

We can construct a new *effective* Hamiltonian by using

$$\mathbf{S}_1 \cdot \mathbf{S}_2 = \begin{cases} -\frac{3}{4} & S = 0 \text{ singlet} \\ \frac{1}{4} & S = 1 \text{ triplet} \end{cases}$$

The new Hamiltonian becomes

$$\mathcal{H}' = \frac{1}{4}(E_S + 3E_T) - (E_S - E_T) \mathbf{S}_1 \cdot \mathbf{S}_2$$

The interesting part is the non-constant spin dependent term. Defining the *exchange constant* J by

$$J := \frac{E_S - E_T}{2} \stackrel{(2.7)}{=} \int \psi_a^*(\mathbf{r}_1) \psi_b^*(\mathbf{r}_2) \mathcal{H} \psi_a(\mathbf{r}_2) \psi_b(\mathbf{r}_1)$$

we define a new effective Hamiltonian

$$\mathcal{H}^{\text{spin}} = -2J \mathbf{S}_1 \cdot \mathbf{S}_2 \quad (2.8)$$

If $J > 0$, $E_S > E_T$ and the triplet state $S = 1$ is favoured. If $J < 0$, $E_T > E_S$ and the singlet state $S = 0$ is favoured.

The above derivation only holds for exactly 2 electrons and the generalization to many-body systems is not trivial. Nevertheless, the Hamiltonian (2.8) motivates the Heisenberg model

$$\mathcal{H} = - \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (2.9)$$

with the factor 2 omitted to prevent counting pairs twice.

2.2.3 Anisotropic Exchange or Dzyaloshinsky-Moriya Interaction

This interaction originates from spin-orbit interactions within one magnetic ion. It can be understood as the exchange interaction between the excited state of one ion

and the ground state of the other.

$$\mathcal{H}_{\text{DM}} = \mathbf{D} \cdot (\mathbf{S}_1 \times \mathbf{S}_2) \quad (2.10)$$

The vector $\mathbf{D}$ vanishes when the crystal field has an inversion symmetry with respect to the centre between the two magnetic ions. However, in general $\mathbf{D}$ may not vanish and then will lie parallel or perpendicular to the line connecting the two spins, depending on the symmetry. The form of the interaction is such that it tries to force $\mathbf{S}_1$ and $\mathbf{S}_2$ to be at right angles in a plane perpendicular to the vector $\mathbf{D}$ in such an orientation as to ensure that the energy is negative. Its effect is therefore very often to cant the spins by a small angle. It commonly occurs in antiferromagnetics and results in a small ferromagnetic component of the moments which is produced perpendicular to the spin axis of the antiferromagnet. The effect is known as *weak ferromagnetism*. It is found in, for example, $\alpha\text{-Fe}_2\text{O}_3$.

2.3 Types of Magnetic Behavior

We discuss magnetic behavior via the classification of magnetic materials. Specifically, we utilize the characteristic dependence of the magnetic susceptibility χ on temperature, applied magnetic field and history:

2.3.1 Diamagnetism

Diamagnetism is defined by

$$\chi^{\text{dia}} < 0 \quad \chi^{\text{dia}} = \text{const.}$$

The classical picture of diamagnetism being an induction effect has already been discussed in 2.1.1. Without discussing proper quantum mechanical treatments like Landau-Diamagnetism in crystals, we focus on phenomenology:

Diamagnetism is a property displayed by *all* materials. However, we only speak of diamagnetism if no other form of magnetism (para- or collective magnetism) is present since it will in general be the weaker effect.

Examples include

- Organic molecules
- Few metals like bismut, zinc, mercury
- Nonmetals like sulfur, iodine, silicon
- *Meissner-Ochsenfeld* effect: Superconductors at $T < T_c$ are perfect diamagnets: $\chi^{\text{dia}} = -1$

As noted, in the presence of e.g. permanent magnetic dipoles, diamagnetism will be a negligible effect and it will not feature throughout this thesis.

2.3.2 Paramagnetism

Typically, one has

$$\chi^{\text{para}} > 0 \quad \chi^{\text{para}} = \chi^{\text{para}}(T)$$

Essentially, paramagnetism is connected to the existence of permanent magnetic dipoles which try to, more or less, orient themselves along an applied auxiliary field $\mathbf{H}$. This competes with thermal motion, hence the temperature dependence of χ^{para} .

In materials, there are two possible origins of such permanent dipoles:

- Localized moments $\rightarrow$ *Langevin paramagnetism*
- Itinerant moments ($\sim$ quasi-free conduction electrons) $\rightarrow$ *Pauli paramagnetism*

Generally, one has

$$\chi^{\text{Langevin}} \gg \chi^{\text{Pauli}} \approx \text{const.}$$

and χ^{Pauli} temperature independent to a first order approximation.

Therefore, only Langevin paramagnetism will feature in this thesis, this however extensively.

2.3.3 Collective Magnetism

The susceptibility in this case is in general a complicated function of the applied field, temperature and the magnetic history of the sample:

$$\chi^{\text{coll.}} = \chi^{\text{coll.}}(T, H, \text{history})$$

Collective magnetism arises due to interactions between the permanent magnetic dipoles. Again, these permanent magnetic dipoles can be either *localized* or *itinerant*

In the context of more standard solid state physics, the interaction in question is the quantum mechanical *exchange interaction* which does not have a classical analogy. As we will discuss later, this view is not sufficient in case of supercrystals comprised of magnetic nanoparticles. Instead, the origin for collective magnetism in our systems, will be the magnetic dipole-dipole interaction between the permanent magnetic moments of the nanoparticles.

Nevertheless, we recall other characteristic features of standard systems with collective magnetism: The exchange interaction leads to a critical temperature T^* below which there exists a *spontaneous magnetization*, i.e. a spontaneous ordering

of dipoles that is not forced externally.

Collective magnetism due to exchange interaction is conventionally divided into three sub-classes:

- Ferromagnetism
- Ferrimagnetism
- Antiferromagnetism

Above the critical temperature T^* , collective magnetic transitions into paramagnetism with the characteristic temperature dependence of the inverse susceptibility sketched in figure 2

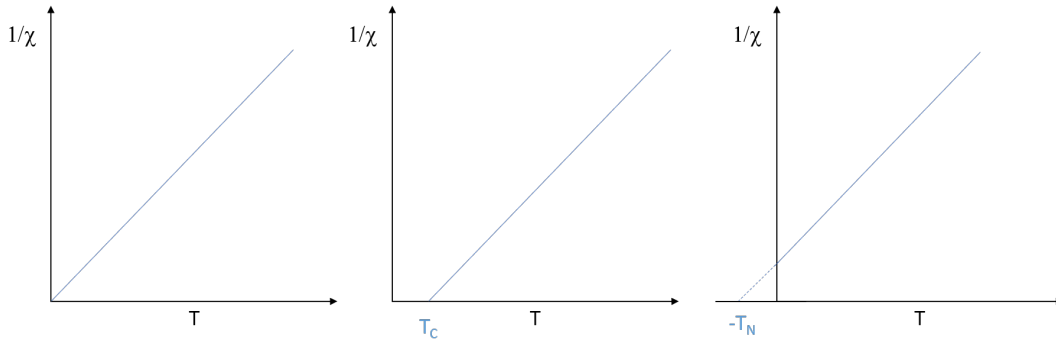


Figure 2: Temperature dependence of the inverse magnetic susceptibility according to the Curie-Weiss law. *Left*: Paramagnetism, *middle*: Ferromagnetism, *right*: Antiferromagnetism

2.3.3.1 Ferromagnetism

In this case the critical temperature is called the *Curie temperature*:

$$T^* = T_C$$

For temperatures $0 < T < T_C$ the permanent moments have a preferential orientation. At $T = 0$, all moments are oriented parallel to each other although $\mu_0 \mathbf{H}_{\text{ext}} = 0$.

2.3.3.2 Ferrimagnetism

In this case the lattice of the system is divided into at least two sublattices A, B with different absolute values of sublattice magnetizations $\mathbf{M}_A, \mathbf{M}_B$ such that

$$\mathbf{M}_A \neq \mathbf{M}_B \quad \text{and} \quad \mathbf{M}_A + \mathbf{M}_B \neq 0 \quad \text{for } T < T_C$$

2.3.3.3 Antiferromagnetism

Here, the critical temperature is called *Néel temperature*:

$$T^* = T_N$$

It is a special case of ferrimagnetism:

$$|\mathbf{M}_A| = |\mathbf{M}_B| \neq 0 \quad \text{and} \quad \mathbf{M}_A + \mathbf{M}_B = 0 \quad \text{for } T < T_C$$

The total magnetization $\mathbf{M} = \mathbf{M}_A + \mathbf{M}_B$ is therefore always zero in the absence of externally applied fields.

2.3.4 Geometrically Frustrated Systems and Spin Glasses

In realistic systems, more than one interaction or energy contribution determines the total energy of the system. The subsequent competition between interactions can lead to complex magnetic behavior that is not trivially following from the constituent interactions.

Even before considering the competition between independent interactions, 2-particle interactions in realistic (super)crystal structures will induce geometric frustration: In many lattices it is not possible to satisfy all the interactions in the system to find *the* ground state. Often this leads to the absence of a single unique ground state but a variety of low energy states.

We refer to this property that the system has no good way to choose which low energy configuration it must adopt as *frustration*. It is important to note that this phenomenon emerges with pretty much any magnetic interaction, including nearest-neighbour interactions such as the antiferromagnetic interaction:

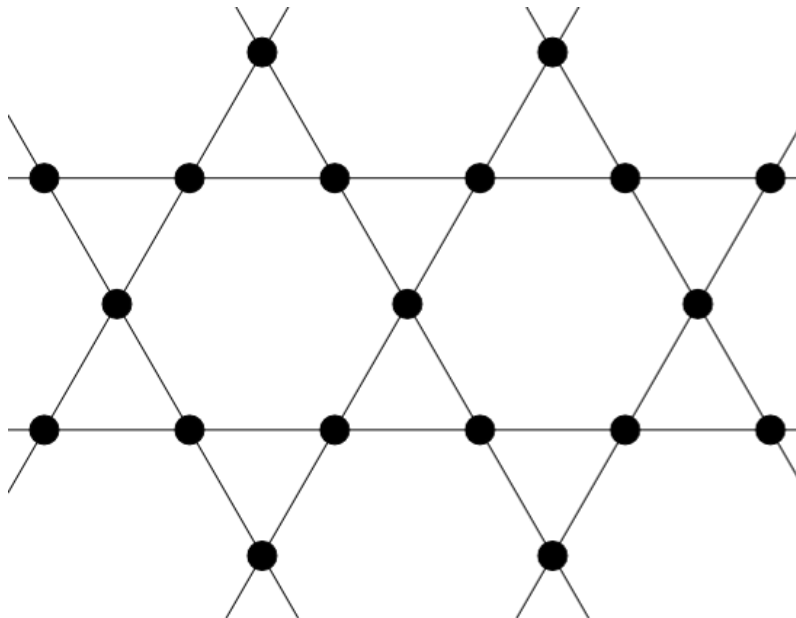


Figure 3: Kagome lattice as an example for a 2D geometry which leads to a frustrated system.

On a square lattice it is easily possible to satisfy the requirement that nearest-neighbour spins must be antiparallel. However on a triangular lattice, things differ: If two adjacent spins are placed antiparallel, the third spin has no good choice

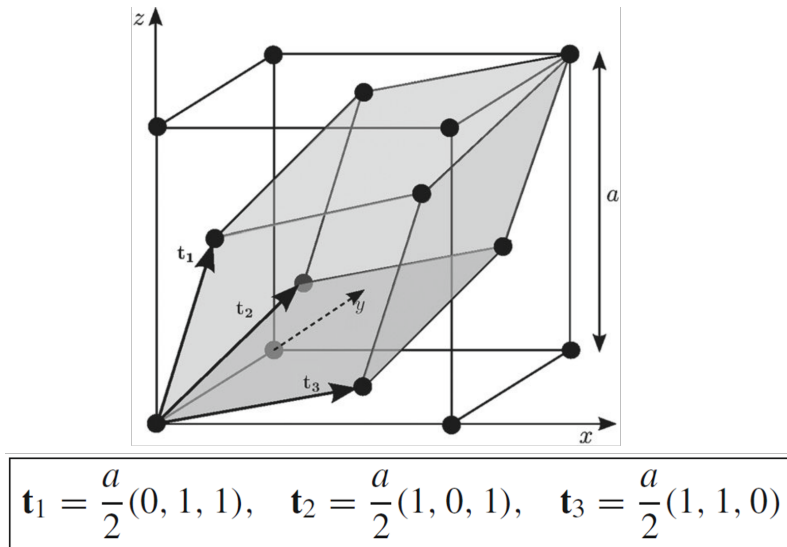


Figure 4: The face-centered-cubic (*fcc*) crystal structure also accounts for geometric frustration and will be heavily featured in future chapters. [7]

between aligning with one or the other spin. Locally, there exists no unique lowest-energy state, but only two low-energy states that are equally unsatisfied.

This minimal frustrated system already shows metastability, hysteresis effects and time-dependent relaxation towards equilibrium, all of which are phenomena absent from the square lattice.

This example relies heavily on the fact that we considered models with low dimensionality, i.e. $\dim < 3$. In three dimensions, frustration would not emerge from a triangular or e.g. *Kagome* lattice, but from a pyrochlore structure in which the magnetic ions occupy a lattice of corner sharing tetrahedra. Here, there is no spin order observable for any temperature, only a classical groundstate with macroscopic degeneracy, sometimes described as *cooperative paramagnetism*.

We can define a spin glass as a random, magnetic system with mixed interactions characterized by a random yet cooperative freezing of spins at a well defined freezing temperature T_f below which a metastable frozen state appears without the usual magnetic long-range ordering.

The decisive term in this definition is *random*. Different types of randomness can be imagined to transform a non-spin glass into a spin glass:

- Site-randomness
- Bond-randomness

Equally important is the presence of competing interactions as previously described. Contributing features include magnetic anisotropy e.g. in amorphous magnets where a random distribution of the *easy-axes* implies random anisotropy.

3 Methods

3.1 Why Do Simulations in Physics

In many cases, models of ideal systems can be explored by theoretical methods, but they do not offer any physical realization so that no comparison to experiment is available. In many other cases, experimental realizations are too complex to be modelled by theoretical methods. In this situation the only possible test for an approximate theoretical solution is to compare with 'data' generated from a computer simulation.

Nuclear reactor meltdowns are a dramatic example: Although we want to know what the results of such events would be, we do not want to carry out experiments. There are also real physical systems which are sufficiently complex that they are not presently amenable to theoretical treatment. An example is the problem of understanding the specific behaviour of a system with many competing interactions and which is undergoing a phase transition. A model Hamiltonian/Hamilton function which is believed to contain all the essential features of the physics may be proposed, and its properties may then be determined from simulations. If the simulation disagrees with experiment, then a new Hamiltonian must be found. An important advantage of simulations is that different physical effects which are simultaneously present in real systems may be isolated and, through separate consideration by simulation, may provide a much better understanding.

The Monte Carlo method has had a considerable history in physics. As far back as 1949 a review of the use of Monte-Carlo simulations using 'modern computing machines' was presented by Metropolis and Ulam [15]. In addition to giving examples they also emphasized the advantages of the method. Of course, in the following decades the kinds of problems they discussed could be treated with far greater sophistication that was possible in the first half of the twentieth century. Nowadays, Monte-Carlo simulation methods have spread into different disciplines that have barely any connection to physics.

3.2 Categories of Monte-Carlo Simulations

A brief overview about Monte-Carlo methods is given. I present additional details about Markov-Chain methods because the research which is presented in this thesis was exclusively done via the Metropolis-Algorithm, the most famous representative of Markov-Chain Monte-Carlo methods.

3.2.1 Monte Carlo in Statistical Physics

Monte-Carlo methods are used throughout many physical and non-physical science branches. In physics, especially statistical mechanics, the following branches are of special interest:

- Monte-Carlo integration
- Importance sampling techniques, specifically Markov Chain methods
 - Local algorithms
 - Non-local algorithms

3.2.2 Markov Chain methods

The concept of Markov chains is central to those Monte-Carlo methods that are the most prominent in physics, especially solid state physics [10].

We define a stochastic process at discrete times labeled consecutively $t_1, t_2, t_3 \dots$ for a system with a finite set of possible states $S_1, S_2, S_3, \dots$, and we denote by X_t the state the system is in a time t . We consider the conditional probability that $X_{t_n} = S_{i_n}$,

$$P(X_{t_n} = S_{i_n} | X_{t_{n-1}} = S_{i_{n-1}}, X_{t_{n-2}} = S_{i_{n-2}}, \dots, X_{t_2} = S_{i_1})$$

given that at the preceding time the system state $X_{t_{n-1}}$ was in state $S_{i_{n-1}}$, etc. Such a process is called a Markov process if this conditional probability is in fact independent of all states but the immediate predecessor, i.e.

$$P(X_{t_n} = S_{i_n} | X_{t_{n-1}} = S_{i_{n-1}})$$

The corresponding sequence of states $\{X_t\}$ is called a Markov chain, and the above conditional probability can be interpreted as the transition probability to move from the state i to state j ,

$$W_{ij} = W(S_i \rightarrow S_j) = P(X_{t_n} = S_j | X_{t_{n-1}} = S_i)$$

We further require that

$$W_{ij} \geq 0 \quad \sum_j W_{ij} = 1$$

as usual for transition probabilities. We may then construct the total probability $P(X_{t_n} = S_j)$ that at time t_n the system is in state S_j as

$$\begin{aligned} P(X_{t_n} = S_j) &= P(X_{t_n} = S_j | X_{t_{n-1}} = S_i) \cdot P(X_{t_{n-1}} = S_i) \\ &= W_{ij} P(X_{t_{n-1}} = S_i) \end{aligned}$$

The master equation conserves the change of this probability with time t (treating time as a continuous rather than discrete variable and writing then $P(X_{t_n} = S_j) =: P(S_j, t)$)

$$\frac{dP(S_j, t)}{dt} = - \sum_i W_{ji} P(S_j, t) + \sum_i W_{ij} P(S_i, t) \quad (3.1)$$

Equation (3.1) can be considered as a 'continuity equation': The total probability is conserved at all times because

$$\sum_j P(S_j, t) \equiv 1 \quad \forall t \in \mathbb{R}$$

Furthermore, all probability of a state i that is 'lost' by transition to state j is gained in the probability of that state, and vice versa.

The Master equation therefore describes the balance of gain and loss processes:

The processes

$$S_j \rightarrow S_{i_1}$$

$$S_j \rightarrow S_{i_2}$$

$$S_j \rightarrow S_{i_3}$$

...

are mutually exclusive. Hence the total probability for a move away from the state j is simply the sum $\sum_i W_{ij} P(S_j, t)$.

We stress that equation (3.1) brings out the basic property of Markov processes:

Basic Property of Markov Processes

The knowledge of the state at time t completely determines the future time evolution.

The main significance is that the importance sampling Monte Carlo process that will feature throughout this thesis via the Metropolis algorithm, can be interpreted as a Markov process if the following is true about the transition probabilities W_{ij} : From now on we require that the transition probabilities satisfy the principle of detailed balance with the equilibrium probability $P_{\text{eq}}(S_j)$:

$$W_{ji} P_{\text{eq}}(S_j) = W_{ij} P_{\text{eq}}(S_i) \quad (3.2)$$

which will be fundamental for the Metropolis algorithm presented in section 3.3.

We already note that (3.2) implies

$$\frac{dP_{\text{eq}}(S_j, t)}{dt} \equiv 0 \quad \forall t \in \mathbb{R}$$

when put into (3.1) because all gain and loss terms cancel exactly. This is elementary for what one would understand by the term 'equilibrium' in context of a system transitioning between states [10].

3.3 The Metropolis Algorithm

From now on, we are only concerned with Monte-Carlo techniques as applied in statistical physics, specifically on-lattice models of systems offering permanent magnetic dipoles that display collective magnetism due to interactions.

In order to illustrate our discussion, we consider the Ising model.

The simple Ising model in zero applied field consists of spins which are confined to the sites of a lattice and which may have only the values $+1$ or -1 . These spins interact with their nearest neighbors on the lattice with interaction constant J ; the Hamiltonian for this model is given by

$$H = -J \sum_{i,j} \sigma_i \sigma_j \quad \sigma_i = \pm 1$$

The Ising model has been solved exactly in one and two dimensions so that Monte-Carlo results in these cases can be directly compared to theoretical expectations.

Next I present the classic Metropolis method.

Configurations are generated from a previous state using a transition probability which depends on the energy difference between the initial and final states. The sequence of states produced follows a time-ordered path, but the time in this case is referred to as 'Monte Carlo time'. For relaxation models, such as we will assume are viable models for magnetization curves of nanoparticle supercrystals, the time-dependent behavior is described by a master equation like (3.1):

$$\frac{\partial P_n(t)}{\partial t} = - \sum_{n \neq m} [P_n(t)W_{n \rightarrow m} - P_m(t)W_{m \rightarrow n}] \quad (3.3)$$

where $P_n(t)$ is the probability of the system being in state n at time t , and $W_{n \rightarrow m}$ is the transition rate for the process $n \rightarrow m$. We again identify the *detailed balance* from (3.2)

$$P_n(t)W_{n \rightarrow m} = P_m(t)W_{m \rightarrow n}$$

as a simple constraint that guarantees an equilibrium being realized as

$$\frac{\partial P_n(t)}{\partial t} \stackrel{\text{equil.}}{=} 0$$

The probability of the n th state occurring in a classical system is given by

$$P_n(t) = \frac{1}{Z} \exp \left(-\frac{E_n}{k_B T} \right) \quad (3.4)$$

where Z is the partition function. Outside of very simple cases like the Ising model discussed here, this expression is very difficult to evaluate, mostly because the partition function, i.e. knowledge about *every possible state and its energy*, is rarely ever known. However, one can avoid this difficulty by generating a Markov chain of states, i.e. generate each new state directly from the preceding state. If we produce the n th state from the m th state, the relative probability is the ratio of the individual probabilities and the denominator, the largely unknown Z cancels. As a result, only the energy difference between the two states is needed, e.g.

$$\Delta E = E_n - E_m$$

The previous idea is possibly the most significant reason why Markov-Chain methods have been so successful in statistical physics because one can circumvent the arduous and ultimately not as interesting task of evaluating the partition function of a large, interacting system.

For the transition rates, any choice that satisfies detailed balance (3.2) is acceptable¹. The first choice of rate which was used in statistical physics is the Metropolis form [15]

$$W_{m \rightarrow n} = \begin{cases} \tau_0^{-1} \exp(-\Delta E/k_B T) & \Delta E > 0 \\ \tau_0^{-1} & \Delta E < 0 \end{cases} \quad (3.5)$$

where τ_0 is the time required to attempt a spin-flip. The way the Metropolis algorithm is implemented can be described by a simple recipe, illustrated in figure 5

Metropolis importance sampling Monte-Carlo scheme (3.6)

1. Choose an initial state
2. Choose a site i
3. Calculate the energy change ΔE which results if the spin at site i is flipped
4. Generate a random number r such that $0 < r < 1$
5. If $r < \exp(-\Delta E/k_B T)$, flip the spin
6. Go to the next site and go to 3.

¹From the derivation from the Master equation (3.1), detailed balance is sufficient but not necessary. It turns out however, that in practice only transition rates that do satisfy detailed balance are regularly used.

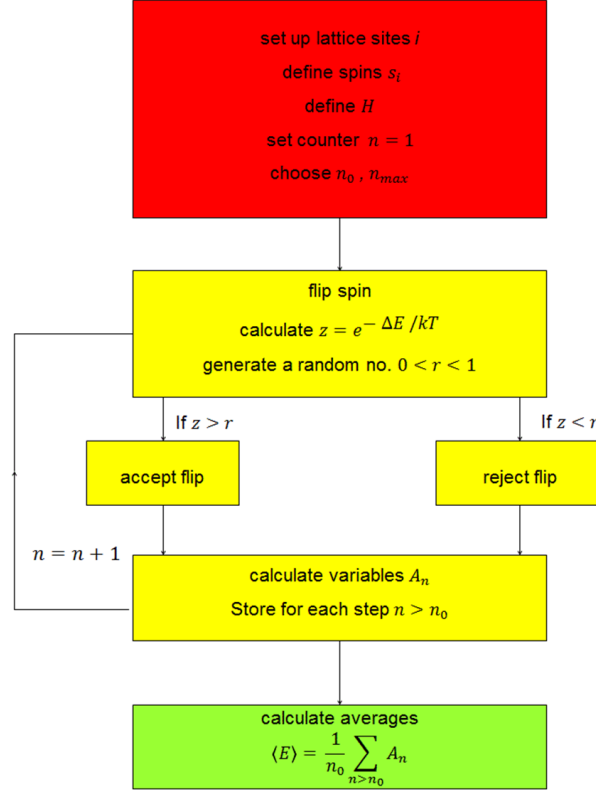


Figure 5: Metropolis Algorithm recipe. The red part is done once in the beginning of a simulation. the yellow part is done in loops, the number of which linearly determines the computation effort and time. The green part is done once after the yellow part has been iterated sufficiently, meaning that one assumes an equilibrium has been reached.

After a certain number of spins have been considered, the properties of the system are determined and added to the statistical average which is being kept. Note that the random number r must be chosen *uniformly* in the interval $[0, 1]$, and successive random numbers should be uncorrelated.

Obviously, this algorithm can be easily modified for use on different on-site models where the total energy of any given configuration can be calculated. Both the required high quality of random numbers and the precise nature of '*flipping*' in the context of the more involved model we consider in the actual research part of this thesis will be thoroughly discussed in section 3.4.

'Monte-Carlo time' is usually measured in terms of Monte Carlo steps per site (MC-S/site) which corresponds to the consideration of every spin in the system once. With the algorithm from figure 5 states are generated with a probability proportional to (3.4) once the number of states is sufficiently large such that any initial transients from the early stages of the iterative loop are negligible. Then the desired averages

$$\langle A \rangle = \sum_n P_n A_n$$

of variables or observables A simply become arithmetic averages over the entire sam-

ple of states which is kept. Note that if an attempted spin-flip is rejected, the old state is counted again for the averaging.

We reiterate that the hallmark of the *Metropolis algorithm* is the specific choice of transition rates given in (3.5). Additional transitions can be imagined like Parallel Tempering which are used in order to accelerate convergence speeds of equilibrium averages for observables or the spin-configuration itself. Also, the choice how to flip spins or, more generally, choose a new configuration for consideration during a subsequent MCS, can be heavily altered compared to figure 5 where one just picks a site randomly.

Both types of modifications lead to algorithms that are not strictly *Metropolis* algorithms, but are still Markov-chain methods if they offer constraints ensuring possible equilibrium, most frequently via enforcing detailed balance [10].

3.4 Quality of Pseudo-random Numbers Picked on a 2-Sphere

The Metropolis algorithm as described in section 3.3 has been used extensively for Ising models where spins are restricted to exactly two states, *up* and *down*. In particular, this implies that it is clear how a local update to achieve a new configuration has to be carried out: Flipping one site to the other state.

In a Heisenberg-like model where each site carries a (super)spin that can assume every position on the 2 dimensional surface of a 3D unit sphere, it is far less clear how an update should be performed. Broadly speaking, 2 types of local updates in a Heisenberg-like on-lattice spin model are possible:

- The possible new states are uniformly distributed on the entire sphere without any bias from position of the original state.
- There is a probabilistic bias which part of the sphere is reachable within one Monte Carlo update.

In this thesis, we will exclusively utilize the second type of spin update. The implementation is depicted in figure 6:

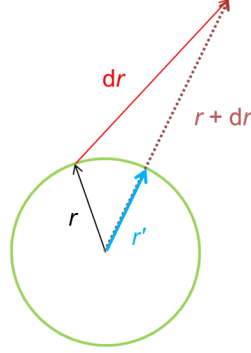


Figure 6: How spins are transformed or 'flipped' during one Monte Carlo step. A random vector $d\mathbf{r}$ (red) is obtained via the Marsaglia method and added to the original $\mathbf{r}$ (black). The result is normalized, yielding $\mathbf{r}'$ (blue). In the end, one has both $\mathbf{r}$ and $\mathbf{r}'$ within a spherical shell with radius 1 (green).

Incremental Transformation

1. Generate an *unbiased*, random vector $d\tilde{\mathbf{r}}$ that is uniformly distributed on a 3D unit sphere and stretch it if necessary with a scalar d_m such that $d\mathbf{r} = d_m d\tilde{\mathbf{r}}$
2. Add this random shift vector to the original $\mathbf{r}$
3. Normalize the result so that $\mathbf{r}'$ is a unit vector.

The generation of a uniform distribution of random vectors on a spherical shell is discussed in section 3.4.1, we can assume for now that we have access to such random vectors of sufficient quality.

This construction ensures that as long as d_m is not too large, $\mathbf{r}'$ is biased towards not deviating too much from $\mathbf{r}$. For example:

$$d_m \lesssim 2 \quad \Rightarrow \quad \mathbf{r}' \neq -\mathbf{r} \quad \forall d\mathbf{r}$$

which means that not every orientation is obtainable within one MCS. The d_m dependence of the $\mathbf{r}'$ distribution is discussed in 3.4.2. In most cases², we choose $d_m = 1$ which means that within 1 MCS there is a heavy bias for new orientations towards the original position. Figuratively, if the original position $\mathbf{r}$ represents the north pole, then $\mathbf{r}'$ is mathematically confined in the northern hemisphere while the equator is only asymptotically obtainable.

²Justification and exemptions are given when needed

3.4.1 Uniformly Distributed Unit Vectors on a $(d - 1)$ -Sphere

For arbitrary dimensions $d \geq 1$ one can generate uniformly distributed vectors on the surface of the corresponding $(d - 1)$ -sphere:

- for $d = 1$: set $\{-1, 1\}$ as boundary of the interval $[-1, 1]$
- for $d = 2$: full circle (as boundary, not area) with radius 1 and origin $(0, 0)$ in cartesian coordinates
- for $d = 3$: spherical shell of the conventional 3-sphere with radius 1 and origin $(0, 0, 0)$ in cartesian coordinates

We can always do this with d random variables which have a Gaussian distribution in an arbitrary interval:

$$\text{Gaussian } (x_1, x_2, \dots, x_d) \quad \mapsto \quad \frac{1}{\sqrt{x_1^2 + x_2^2 + \dots + x_d^2}} \cdot \begin{pmatrix} x_1 \\ x_2 \\ \vdots \\ x_d \end{pmatrix}$$

This d -vector will be uniformly distributed on the surface of a d -sphere.

For the special case $d = 3$ that we are interested in however, there is a more elegant method by Marsaglia [13] which requires only 2 instead of 3 random numbers:

Marsaglia Method

1. Pick a and b from independent uniform distributions on $(-1, 1)$
2. Reject points for which $a^2 + b^2 \geq 1$
3. From the remaining points
 - $x = 2a\sqrt{1 - a^2 - b^2}$
 - $y = 2b\sqrt{1 - a^2 - b^2}$
 - $z = 1 - 2(a^2 + b^2)$

The vectors $\begin{pmatrix} x \\ y \\ z \end{pmatrix}$ then have a uniform distribution on the surface of a unit sphere as needed in our spin-update scheme.

3.4.2 Examples of Vector Distribution

Next we demonstrate the influence of the length of the test vector d_m onto the vector distribution of a modified vector after a certain number of steps according to our recipe in figure 6.

The following graphs correspond to this scenario:

1. 10^6 unit vectors are initialized identically with orientation $(1, 0, 0)$
2. Choose $d_m = 2$.
3. With independent Marsaglia random vectors, perform the transformation.
 - once $\rightarrow$ graphs in figures 7, 8, 9.
 - 1000 times $\rightarrow$ graphs in figures 10, 11, 12.
4. Record the component distribution of the resulting vectors after all transformations are done.
5. Fill histograms with bins of width 0.002.

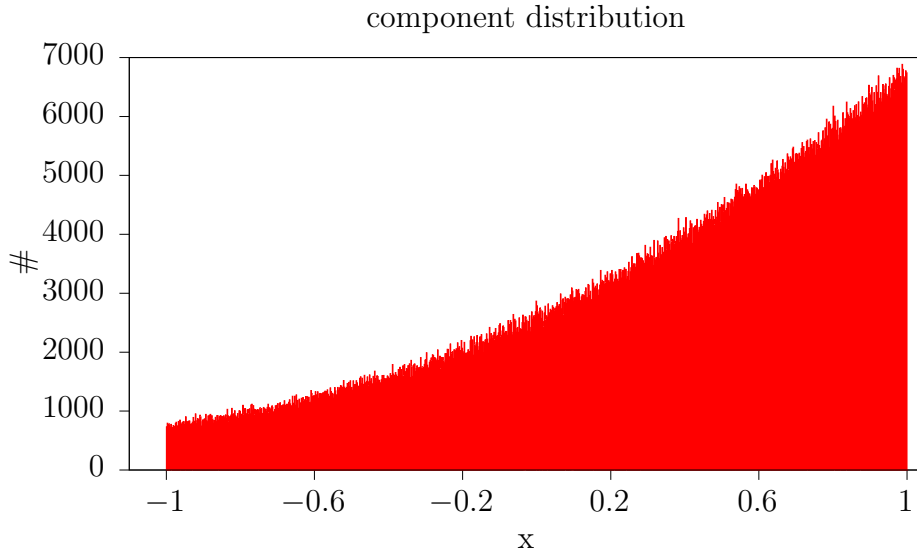


Figure 7: Distribution of x-components after 1 loop

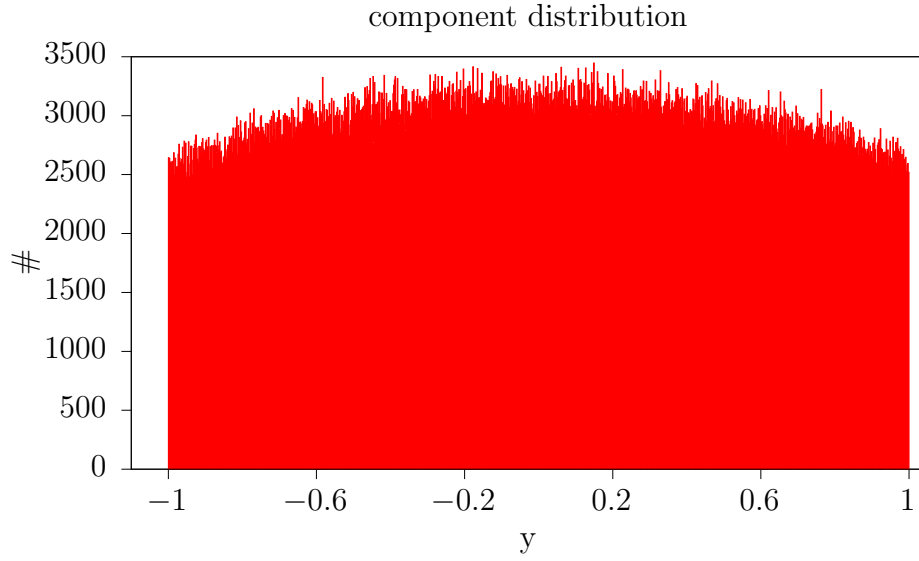


Figure 8: Distribution of y-components after 1 loop

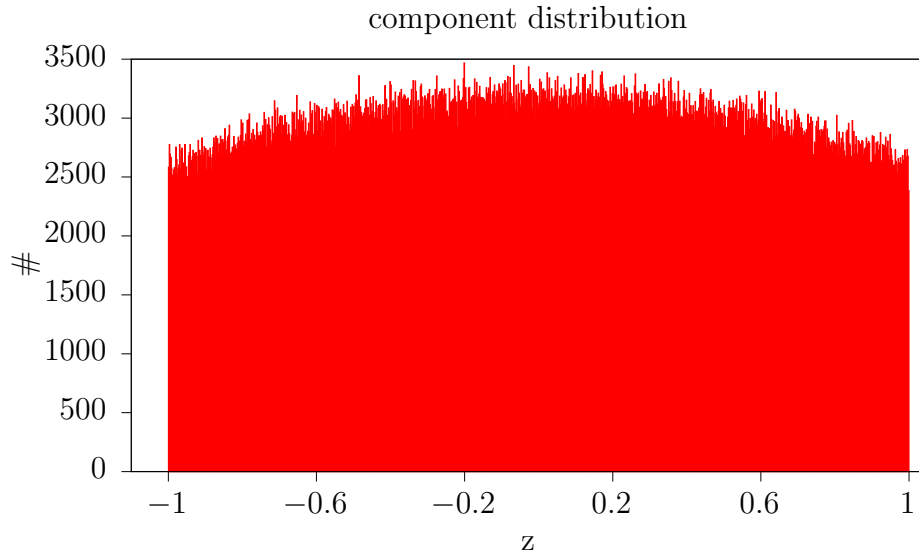


Figure 9: Distribution of z-components after 1 loop

We observe that the x-component covers the complete interval $(-1, 1)$, but the distribution is skewed in favor of the original orientation $x \rightarrow 1$. The y- and z-components follow identical distributions, centered around 0. Note that the integrated y- and z-components are less than the x-component because one 'flip' cannot undo the non-uniform start distribution around $(1, 0, 0)$.

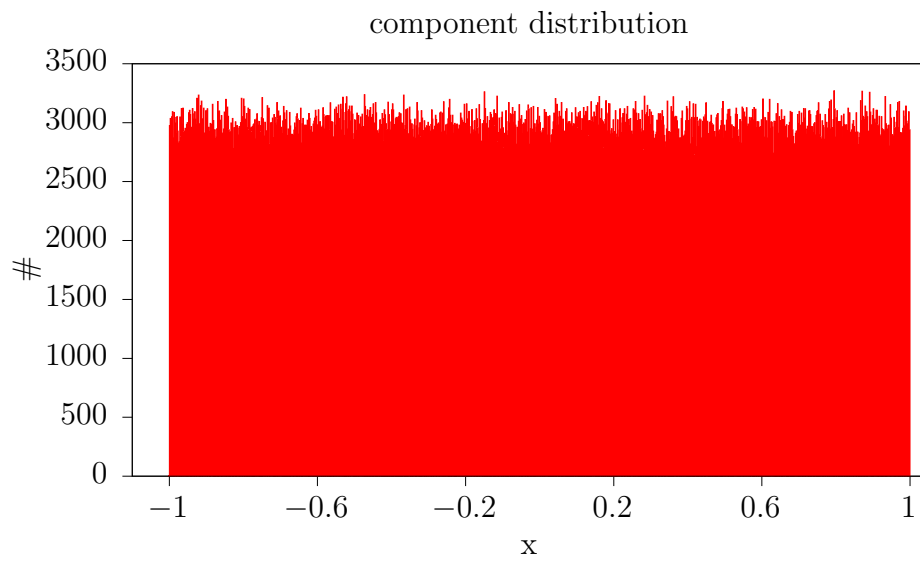


Figure 10: Distribution of x-components after 1000 loops

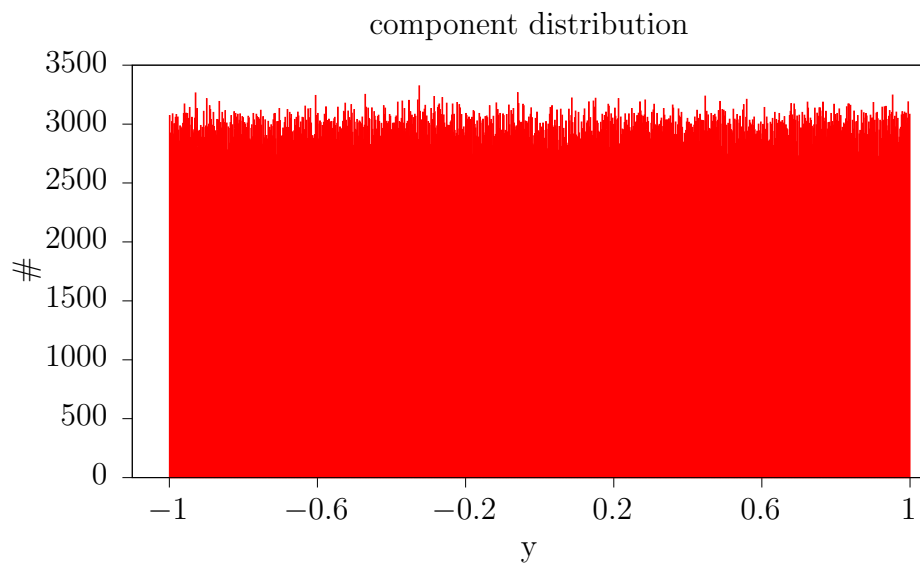


Figure 11: Distribution of y-components after 1000 loops

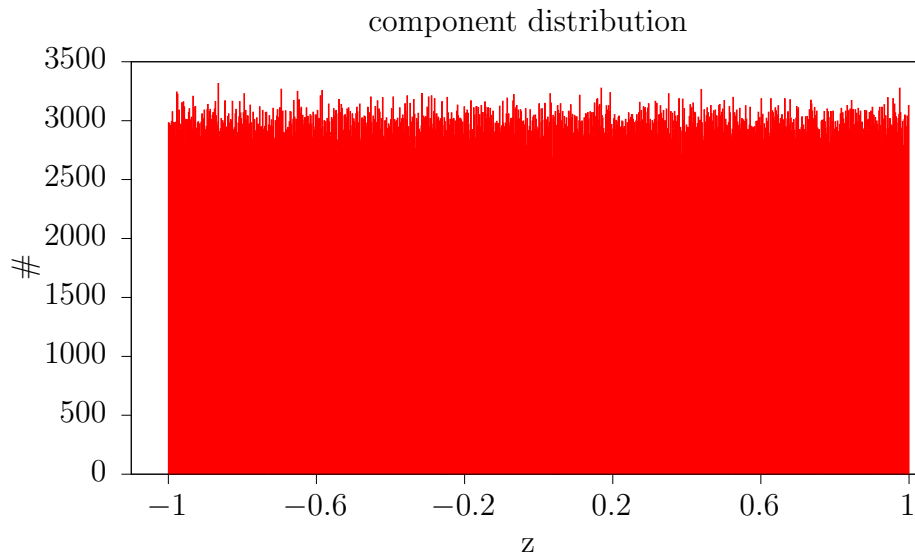


Figure 12: Distribution of z -components after 1000 loops

In this second set of graphs, we see that a sufficient number of transformation steps produces a distribution that is indistinguishable from a uniform distribution.

3.4.3 MC simulations and Ergodicity

We give a definition of ergodicity in the language of Monte-Carlo simulations on realistic machines with finite precision:

Definition: Ergodicity

In the context of a MC simulation, ergodicity means that the implementation of the algorithm ensures that all points in the simulated phase space of the system are eventually visited after a finite number of simulation steps.

Because of the finite nature of the precision of the machine, the phase space is also finite, albeit quite large. This is a subtle difference to the original definition of ergodicity in an uncountable phase space where each point merely is arbitrarily closely matched after sufficient, but also finite time.

In this thesis, we are interested in measurements taken at thermal equilibrium and therefore ergodicity is generally *assumed* on the level of microstates. This assumption is also referred to as *ergodic hypothesis*

Ergodic Hypothesis

Thermodynamic systems evolve in a way that all energetically allowed regions in phase space are covered. The time that the trajectory stays in a particular region of phase space is proportional to the phase-space volume of this region.

The consequence of the above is that a necessary condition for our implementations is that one must choose a combination of d_m and number of MCS that allows any microstate evolving to any other microstate. Additionally, on average there must not be any bias left in favor of phase space regions 'closer' to the original region. The last point implies that a distribution like in figure 10 is fine while a distribution like figure 7 is in violation of the ergodic hypothesis and thus cannot correspond to a system at thermal equilibrium.

4 Ferromagnetic Nanoparticles and Supercrystals in the Non-interacting Limit

The main goal of this thesis is to further the knowledge of dipolar magnetic systems. Because the interactions are long-range, a theoretical treatment is presently impossible. Comparing the simulation results with experiments will at least provide answers to questions like 'Do we know which energies are important' and 'Is the single-domain approximation valid'?

4.1 Theory I

Throughout section 4 we will not need to consider any interactions between the magnetic moments of the nanoparticles. The physical properties we simulate here are therefore comparatively easy to treat rigorously. This will be discussed next.

4.1.1 Stoner-Wohlfarth Model

Two energies will be considered in this chapter.

- The magnetostatic energy of one nanoparticle magnetic moment ('superspin') $\mathbf{m}$ in an external magnetic field

$$E_m = -\mathbf{m} \cdot \mathbf{B} = -m B \cos(\theta)$$
- The magnetocrystalline anisotropy energy $E_a = K V \sin^2(\delta)$

which is quantified according to the Stoner-Wohlfarth model for uniaxial anisotropy constants.

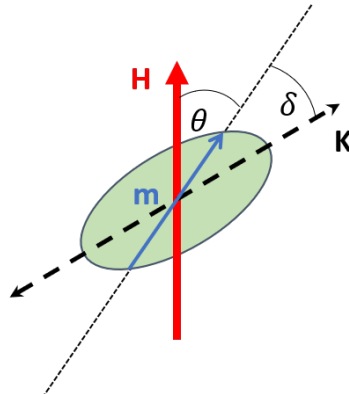


Figure 13: Geometry of the uniaxial Stoner-Wohlfarth model

$$E = E(\theta, \delta) = -mB \cos(\theta) + KV \sin^2(\delta)$$

or, in order to use the experimentally more common applied field strength $\mathbf{H}$:

$$E = -\mu_0 H m \cos(\theta) + KV \sin^2(\delta)$$

4.1.2 Isotropic Paramagnet

First, we will ignore the second energy and try to recover the both analytically and experimentally well-known result for the magnetization curve of such a paramagnet in an external field. In nature, the vanishing of any magnetocrystalline anisotropy is not usual. The best analogy would be a material where the saturation magnetization is rather high compared to the anisotropy constant.

We define the total energy of the system being given as

$$E = -\mathbf{m} \cdot \mathbf{B}$$

where the modulus $|\mathbf{m}| = m$ is constant and $\mathbf{B} = B\hat{\mathbf{e}}_z$. When calculating the expectation value of

$$m_z = m \cos \theta$$

in the canonical ensemble, we therefore have to consider all possible orientations of $\mathbf{m}$ on $\mathcal{S}^2$, the surface of the 3-sphere, according to:

$$\begin{aligned} \langle m_z \rangle &= \frac{1}{Z} \int_{\mathcal{S}^2} m_z \exp \left(\frac{\mathbf{m} \cdot \mathbf{B}}{k_B T} \right) d^2r \\ \text{with } Z &= \int_{\mathcal{S}^2} \exp \left(\frac{\mathbf{m} \cdot \mathbf{B}}{k_B T} \right) d^2r \\ \varphi\text{-Symmetry} \Rightarrow \langle m_z \rangle &= m \frac{\int_0^\pi \exp \left(\frac{mB \cos \theta}{k_B T} \right) \sin \theta \cos \theta \, d\theta}{\int_0^\pi \exp \left(\frac{mB \cos \theta}{k_B T} \right) \sin \theta \, d\theta} \end{aligned}$$

where we used standard spherical coordinates (r, θ, φ) and $z = r \cos \theta$. Defining:

$$x := \frac{mB}{k_B T} \quad v := \cos \theta$$

we get the easily solvable

$$\langle m_z \rangle = m \frac{\int_{-1}^1 v e^{xv} dv}{\int_{-1}^1 e^{xv} dv} \tag{4.1}$$

The magnetic moments are indistinguishable. We therefore get for the total magnetization M if n is the number *density* of magnetic moments, i.e. the number of magnetic moments, each with modulus m , per Volume

$$M(T, B) = n \langle m_z \rangle = M_{\max} L \left(\frac{mB}{k_B T} \right) \tag{4.2}$$

with the solution of (4.1), the *Langevin function* L

$$L(x) = \frac{1}{\tanh(x)} - \frac{1}{x} = \frac{x}{3} + O(x^3)$$

and the saturation magnetization, read maximum achievable magnetization

$$M_{\max} = n m$$

For small fields B , the magnetic susceptibility can be written as

$$\chi = \frac{M}{H} \sim \frac{\mu_0 M}{B} = \frac{n \mu_0 m^2}{3 k_B T} \quad (4.3)$$

In particular, we get $\chi \propto 1/T$ which is known as Curie's law and is an important hallmark of paramagnetic systems or their analogies. We can rewrite the Curie law as

$$\chi = \frac{C}{T} \quad (4.4)$$

with the Curie constant C which we here expect to be

$$C = \frac{n \mu_0 m^2}{3 k_B} \quad (4.5)$$

or, in case that we have particles of finite volume V_{part} and saturation magnetization *per particle volume* M_s :

$$\begin{aligned} M_{\max} &= n M_s V_{\text{part.}} \\ C &= \frac{n \mu_0 (M_s V_{\text{part.}})^2}{3 k_B} \end{aligned} \quad (4.6)$$

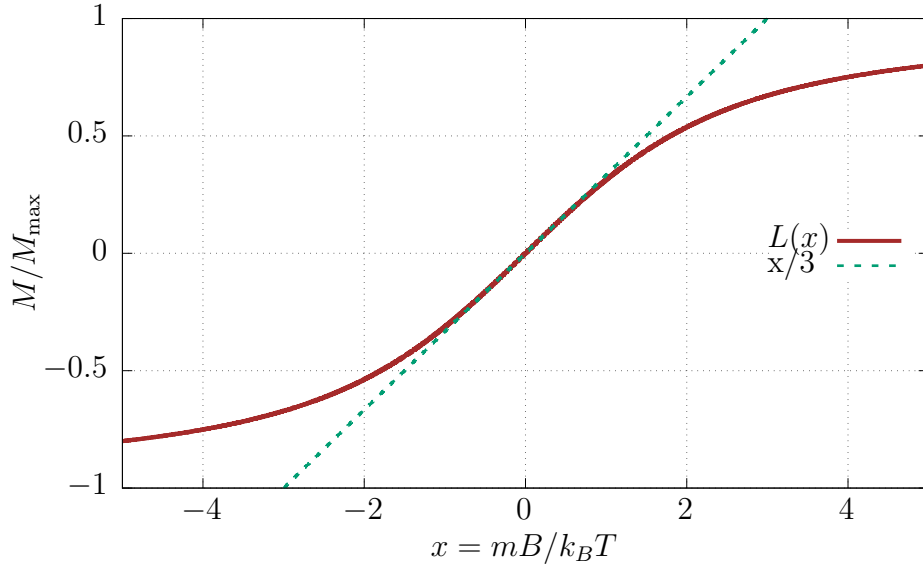


Figure 14: Langevin function and small-field approximation

4.1.3 Potential Landscape with Non-vanishing Anisotropy

We will now also include the magnetocrystalline anisotropy energy. It is important to note that we thereby have introduced at least two additional parameters: The magnitude of the anisotropy constant K and the distribution of the easy axes $\mathbf{k}$ where E_a is minimal if $\pm\mathbf{m} \parallel \mathbf{k}$.

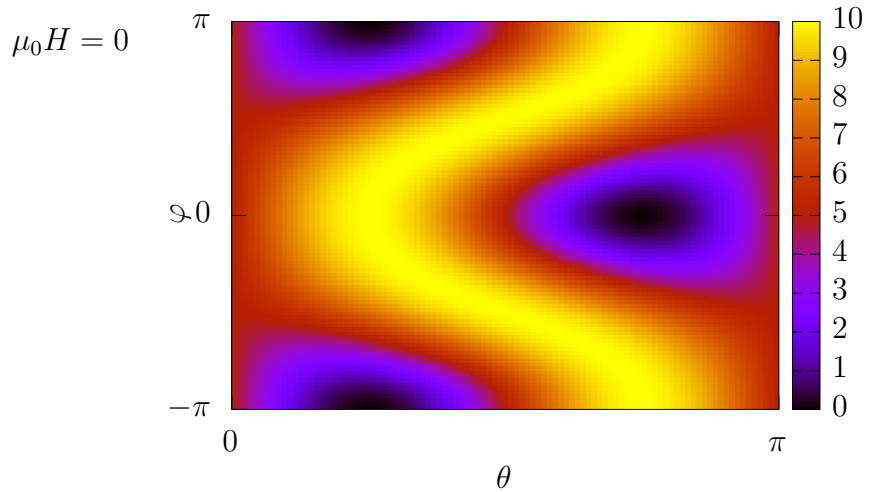


Figure 15: Potential landscape in spherical coordinates without external field. Energy in arbitrary units, following the given parameter set.

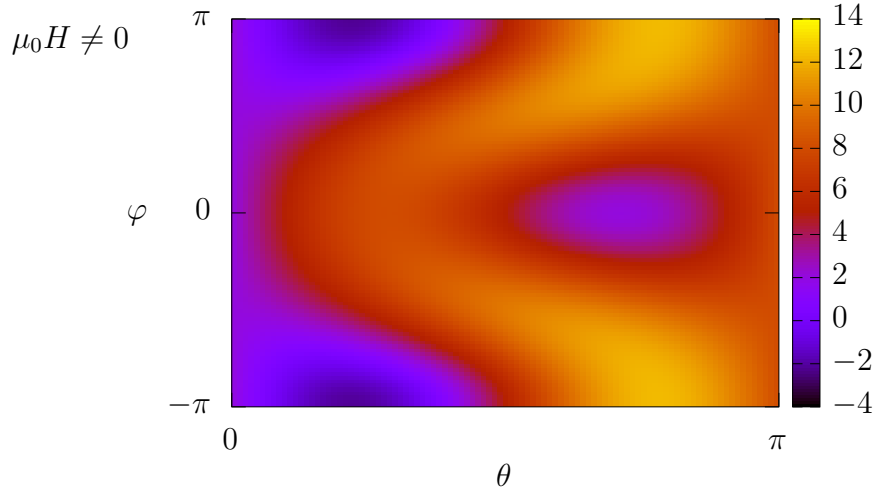


Figure 16: Potential landscape in spherical coordinates with external field

Figures 15 and 16 show the potential landscape for one superspin orientation in one setting of parameters in spherical coordinates (θ, φ) :

- $KV = 5 \quad mB = 1.5 \quad (\text{arbitrary, non-physical units})$
- $\hat{\mathbf{B}} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \quad \hat{\mathbf{k}} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ 0 \\ -1 \end{pmatrix}$
- $E = E(\theta, \varphi) = KV \{1 - \cos^2[\delta(\theta, \varphi)]\} - mB \cos \theta$
- $\cos[\delta(\theta, \varphi)] = \frac{1}{\sqrt{2}}(\sin \theta \cos \varphi - \cos \theta)$

Figures 17 and 18 show profiles for specific choices of φ . Note that any external field breaks the π -periodicity in θ into a 2π -periodicity.

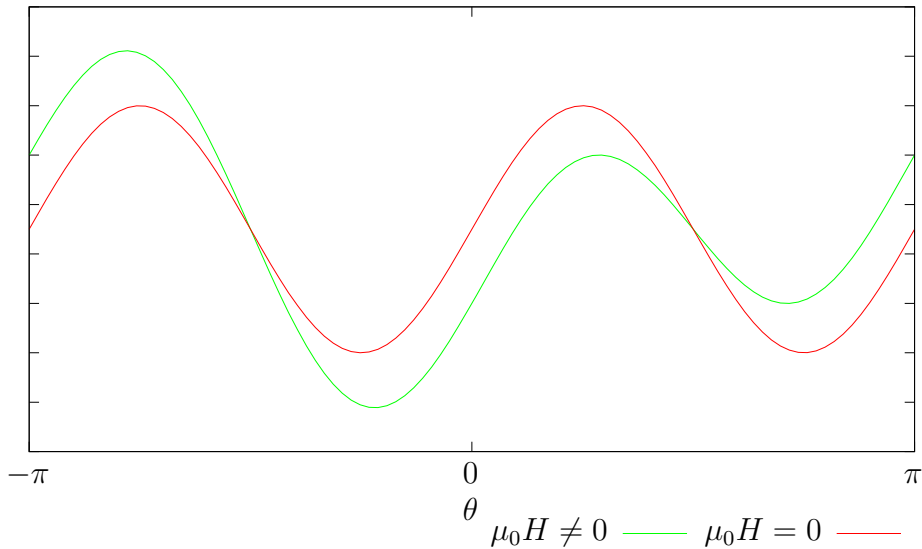


Figure 17: Profile of potential in figures 15, 16 for $\varphi = 0$

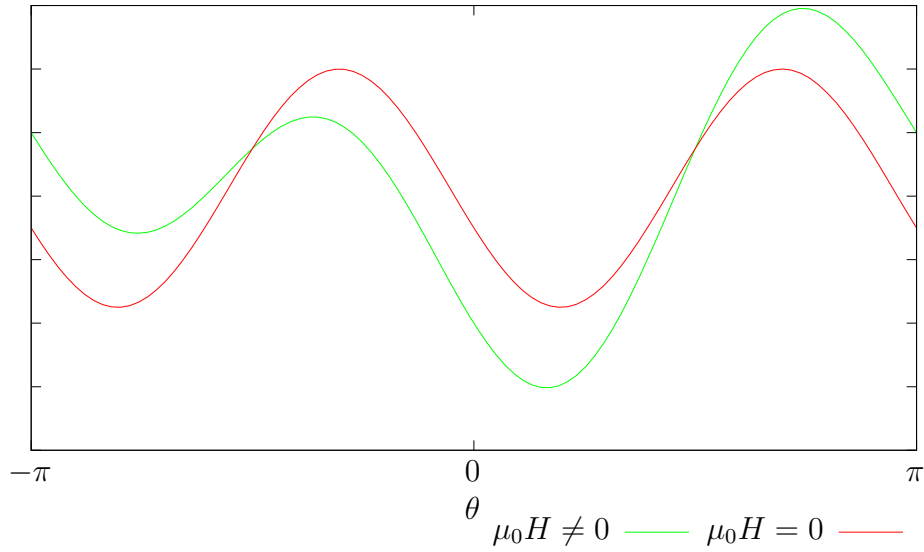


Figure 18: Profile of potential in figures 15, 16 for $\varphi = 3\pi/4$

4.1.4 ZFC-FC curves

A very common experimental procedure is recording the *Zero-field-cooled* (ZFC) and *Field-cooling* (FC) curves. The sample undergoes (at least partly) the following path and typically shows a magnetization curve depending on T as given in figure 19.

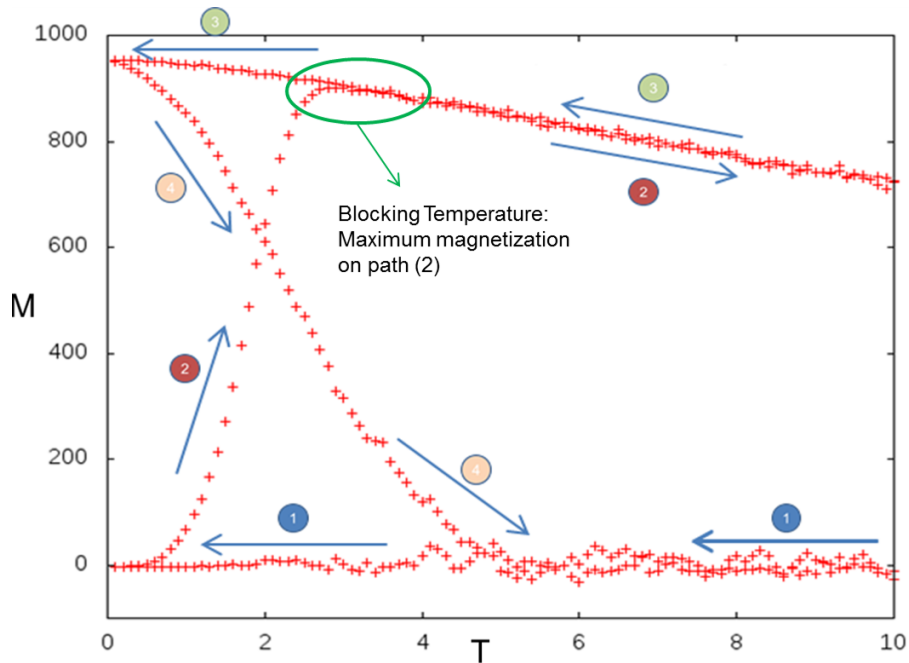


Figure 19: Idealized $M(T)$ curve with arbitrary units. Path 2, 3 are the ZFC, FC branches

Standard Temperature-Sweep Magnetometric Experiment

1. The system is randomly generated at a sufficiently large starting temperature with $\mu_0 H = 0$ and cooled down
2. System is heated with a constant $\mu_0 H$ applied (*ZFC curve*)
3. without change in the applied field, system is cooled again (*FC curve*)
4. at $H = 0$, is now heated again

The fourth branch is most often omitted whereas the first may not have a name but is still essential for branches 2 and 3.

One of the characteristic quantities of these magnetization curves is the *Blocking temperature* T_B which can (roughly) be defined as the temperature where during the ZFC-curve the magnetization reaches its maximum. The term blocking refers to the fact that above T_B , the system is expected to behave like a paramagnet. Since the involved magnetic moments are the nanoparticle superspins, this behavior is referred to as *Superparamagnetism*.

4.1.5 ac-Susceptibility

In case of ideal monodisperse nanoparticles as we mostly consider them, the relaxation time of particles obeys the Néel formula

$$\tau \simeq \tau_0 \exp\left(\frac{\theta}{T}\right) \quad \frac{\theta}{T} \equiv \frac{KV}{k_B T}$$

with volume V and uniaxial anisotropy constant K

we furthermore recall the static susceptibility (Langevin susceptibility) χ_0

$$\chi_0 = \frac{(M_s V)^2}{3k_B T}$$

with saturation magnetization M_s . Then, for an applied ac field, one finds

$$M(t) = H_0 (\chi' \cos \omega t + \chi'' \sin \omega t)$$

This implies that the magnetization will try to follow the applied field but would do so with a phase lag.

We present two cases of an ac-susceptibility experiment:

1. Apply a saturating field onto the system and have it relax before applying the sinusoidal H_{ac}
 $\rightarrow \chi_{SAT}$

2. Have the sample cooled in zero-field and then switch on H_{ac}
 $\rightarrow \chi_{ZFC}$

In the first case, one assumes that after switching off the saturating field, any single particle will undergo the following exponential decay (Néel relaxation)

$$M(t) = M_{eq} e^{-t/\tau}$$

This leads to the following real and imaginary part of the susceptibility in the model of Néel relaxation after saturation [1]:

$$\begin{aligned}\chi'_{SAT}(\omega) &= \chi_0 \frac{1}{1 + (\omega\tau)^2} \\ \chi''_{SAT}(\omega) &= \chi_0 \frac{\omega\tau}{1 + (\omega\tau)^2}\end{aligned}$$

The second case also exhibits Néel relaxation as basis for the magnetization dynamics. However, an important difference is that as soon as any field is applied to the zero-field cooled sample, a net magnetization occurs instantaneously (as described in 4.1.4) which then leads to a temperature-independent nonzero contribution to χ_{ZFC} .

The real-time result for the ZFC susceptibility χ becomes [1]

$$\chi_{ZFC}(t) = \frac{M_s^2}{3K} \left[1 + \frac{KV}{k_B T} (1 - e^{-t/\tau}) \right]$$

and a Fourier transformation yields

$$\chi'_{ZFC}(\omega) = \frac{M_s^2}{3K} \left[1 + \frac{KV}{k_B T} \frac{1}{1 + (\omega\tau)^2} \right] \quad (4.7)$$

$$\chi''_{ZFC}(\omega) = \frac{M_s^2}{3} \frac{V}{k_B T} \frac{\omega\tau}{1 + (\omega\tau)^2} \quad (4.8)$$

Only simulation results for the ZFC-ac susceptibility and the first setup will be presented. In future research however, the alternative setup may be of particular interest if samples with non-vanishing interactions are considered. Via eliminating $\omega\tau$ we obtain the following relationship between χ' , χ'' :

$$\left(\chi' - \frac{\alpha(2 + \sigma)}{2} \right)^2 + \chi''^2 = \left(\frac{\alpha\sigma}{2} \right)^2 \quad (4.9)$$

where

$$\alpha \equiv \frac{M_s^2}{3K} \quad \sigma \equiv \frac{KV}{k_B T}$$

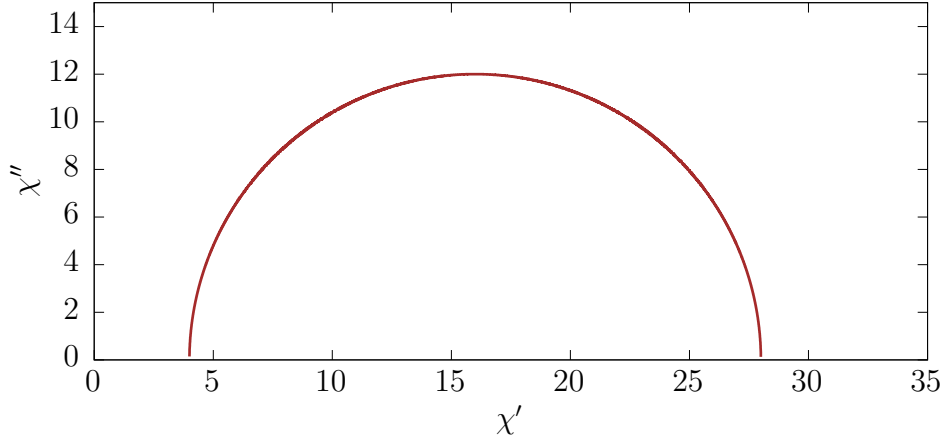


Figure 20: Theoretical result for non-interacting Cole-Cole plot with $c = 16$, $r = 12$

we obtain the formula of a semi-circle.

This means that the so-called Cole-Cole plot of real vs. imaginary part of χ_{ac} is a perfect semi-circle in the first quadrant with its center coordinates

$$(c, 0) \equiv (\alpha(2 + \sigma)/2, 0)$$

and radius

$$r = \alpha\sigma/2$$

Furthermore, both the anisotropy constant and saturation magnetization can be extracted from such a circle plot

$$KV = 2k_B T \frac{r}{c - r}$$

$$M_s = \sqrt{6 \frac{k_B T}{V}} \sqrt{r}$$

4.2 Simulation results

Throughout the following chapters, we will almost exclusively consider *maghemite* (γ -Fe₂O₃) [2] nanoparticles with uniformly distributed easy axes distribution and realistic parameters as they are given in nature.

4.2.1 Isotropic Superparamagnetism

We study the limit of vanishing magnetocrystalline anisotropy for magnetic nanoparticles and aim to recover the behavior of an isotropic paramagnet.

To this end, at constant temperatures several magnetic fields are applied to the nanoparticles and the magnetization is recorded. Results are given in figure 21.

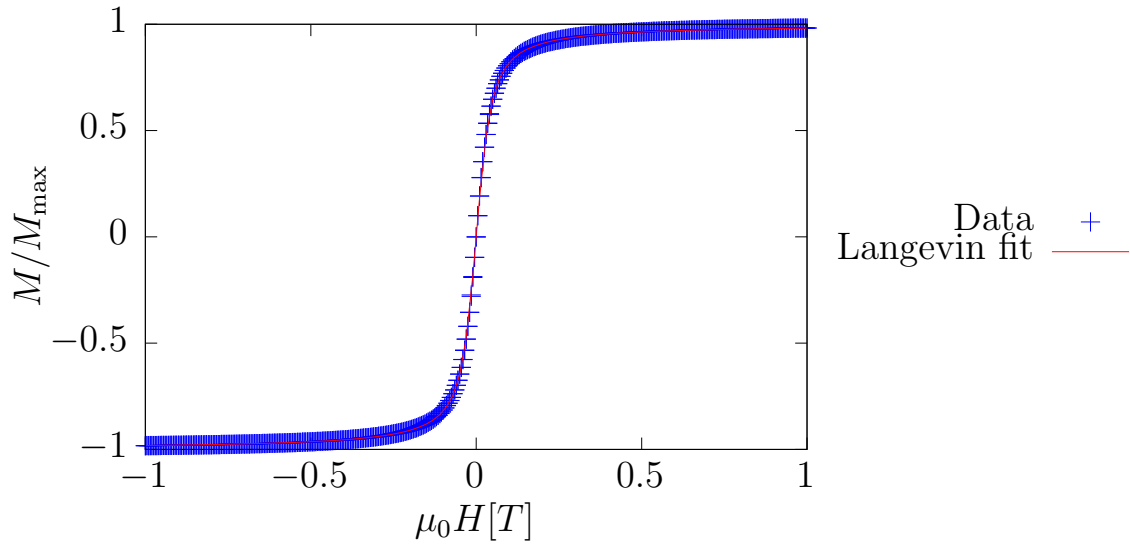


Figure 21: Simulation results for the isotropic paramagnet and the fit of the Langevin expectation

Fitting the theoretically expected Langevin function lets us recover the saturation magnetization that we implemented perfectly. We have therefore shown that our Monte-Carlo approach is in accordance with both theory and experiment.

4.2.2 ZFC-FC Curves

Several magnetometric simulations were carried out where appropriate temperature intervals were swept in the presence of static magnetic fields.

4.2.2.1 High-Temperature Behavior

We are interested in the behavior of the ZFC curve well above T_B where theory predicts the sample to behave like an isotropic paramagnet.

In particular, one expects the magnetization to follow a Curie-Weiss law

$$M(T) = M_\infty + \frac{C}{T - T_C}$$

with both $M_\infty = 0$ and $T_C = 0$ because anything other would indicate a non-vanishing permanent magnetization after the zero-field cooling or (anti-)ferromagnetic behavior. Also, the Curie constant C can be compared to equation (4.6)

There is nothing in our model allowing for either. Specifically, any $T_C \neq 0$ would indicate interactions among our superspins that would have to originate from errors in the implementation or a poor quality in the utilized pseudo-RNG.

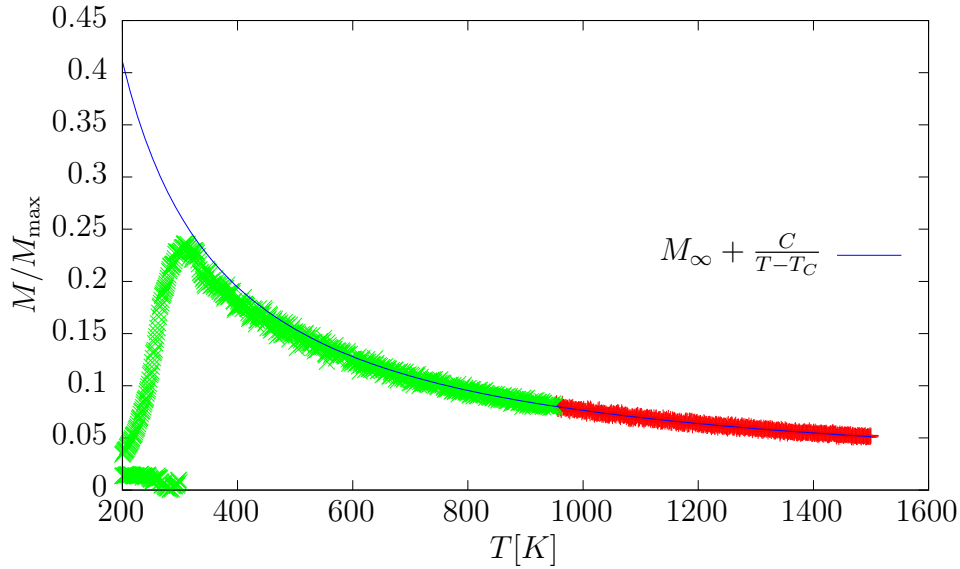


Figure 22: Fitting Curie-Weiss law onto the high-temperature tail of the ZFC-curve at $\mu_0 H = 2 \text{ mT}$

We find that our computer experiment is in accordance with the expectation.

4.2.3 Influences on the Blocking Temperature

When doing a simulation, we still have to specify the following parameters:

- number of particles
- modulus of the test vector (see figure 6)
- number of Monte-Carlo steps (distributed among relaxation- and averaging-loops) per measurement point
- magnitude and direction of the applied magnetic field $\mu_0 H$
- distribution and magnitude of uniaxial anisotropy constants

Each of these parameters may influence the observed blocking temperature in our computer experiment. (As it turns out, all of the above do so, at least in the case of interacting, periodic systems which are discussed later).

4.2.3.1 Particle Number

Figure 23 shows simulations of the ZFC-curve for a different number of superspins in the sample but otherwise same parameters:

- Test vector modulus $|\mathbf{d}_m| = 1$
- Monte-Carlo steps per point: $2000^{(\text{relaxation})} + 10^{(\text{averaging})}$

- applied field $\mu_0 H = 15$ mT

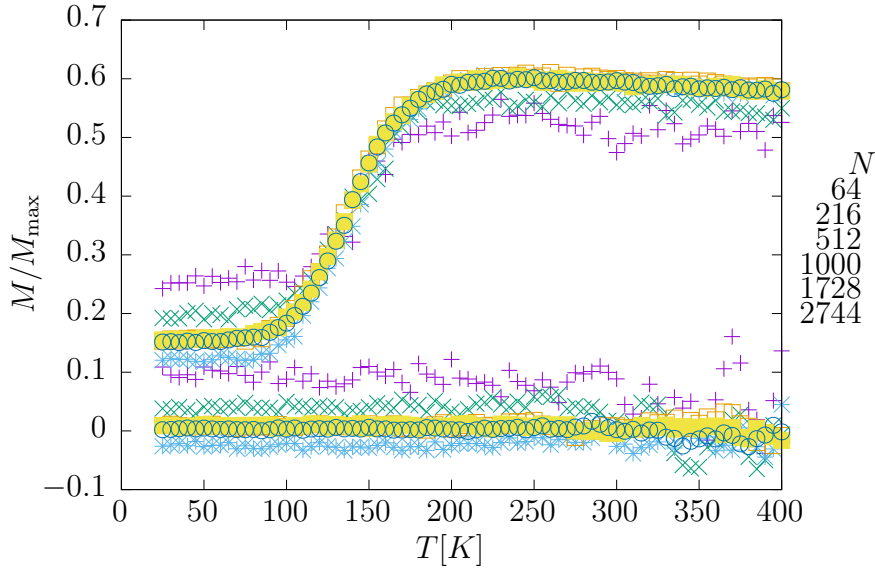


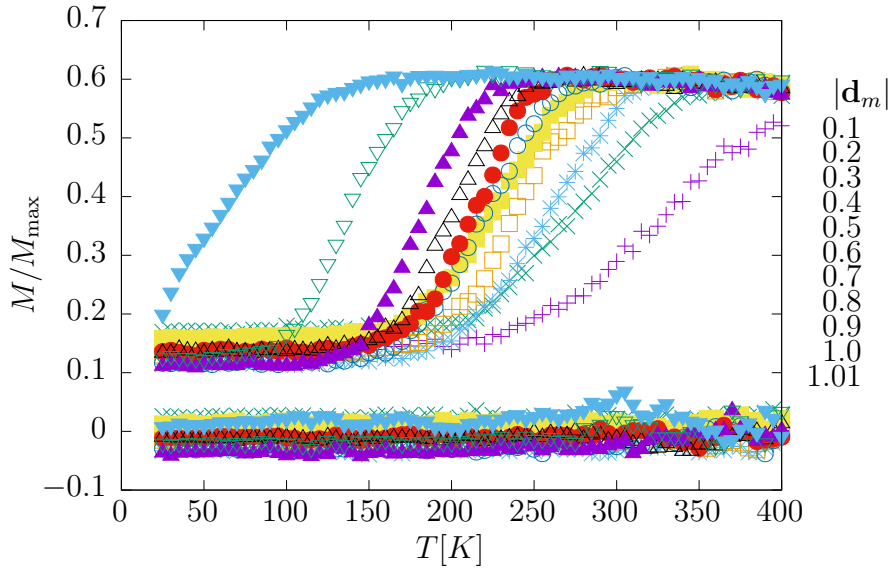
Figure 23: ZFC curves with varying number of particles

We see that for low N , the statistical quality of the simulation is poor since for example no T_B can be extracted well. Other than that, we do not observe different dynamics for greater N , but less noisy data. This is to be expected because we do not consider any interaction between particles in this chapter.

4.2.3.2 Test Vector

Figure 24 shows simulations of the ZFC-curve for different test vectors moduli in the sample but otherwise same parameters:

- number of magnetic moments: 1000
- Monte-Carlo steps per point: $2000^{(\text{relaxation})} + 10^{(\text{averaging})}$
- applied field $\mu_0 H = 15$ mT


 Figure 24: ZFC curves with varying modulus of test vector $|\mathbf{d}_m|$

This is probably the most involved parameter study in 4.2.2. The analysis of the component distribution for the transformed vectors as demonstrated in 3.4.2 shows for all d_m that were considered here, that after 2000 MCS the vectors would be uniformly distributed *in the absence of any potential*. Yet we see dramatically different behaviour in terms of observed T_B . We see an increase of T_B for decreasing d_m , which is intuitive because a smaller d_m corresponds to a slower dynamic of any individual nanoparticle whereas the speed of temperature change is constant in all 11 plots shown. Especially the non-linear behaviour around $d_m = 1$ however is only plausible if we recall this geometric insight: For this test-vector length, a jump 'from one pole to the equator' is barely possible. This is precisely the distance between the minima of the Stoner-Wohlfarth model. This means that for $d_m > 1$, a jump from the energetically unfavourable equilibrium state to the favourable one is possible (though still not very likely) within one Monte-Carlo update, which seems counterintuitive for systems at low temperature. For smaller d_m where this quick channel is completely forbidden, which leads to significantly slower dynamics. The geometric significance of the test-vector length must thus be taken into account when designing simulation parameters for other energy landscapes. A balance must be found between the economy of wasting too much computation time in uneventful parts of phase space ($\leftrightarrow d_m$ chosen too low) and the phenomenon of spins leap-frogging potential landscapes at low temperatures ($\leftrightarrow d_m$ chosen too big).

4.2.3.3 Number of Monte-Carlo Steps per Measurement

Figure 25 shows simulations of the ZFC-curve for different numbers of *relaxation*-Monte-Carlo steps per measurement point but otherwise same parameters, including *averaging* Monte-Carlo steps:

- number of magnetic moments: 1000
- Test vector modulus $|\mathbf{d}_m| = 1$
- Monte-Carlo *averaging* steps per point: 10
- applied field $\mu_0 H = 15$ mT

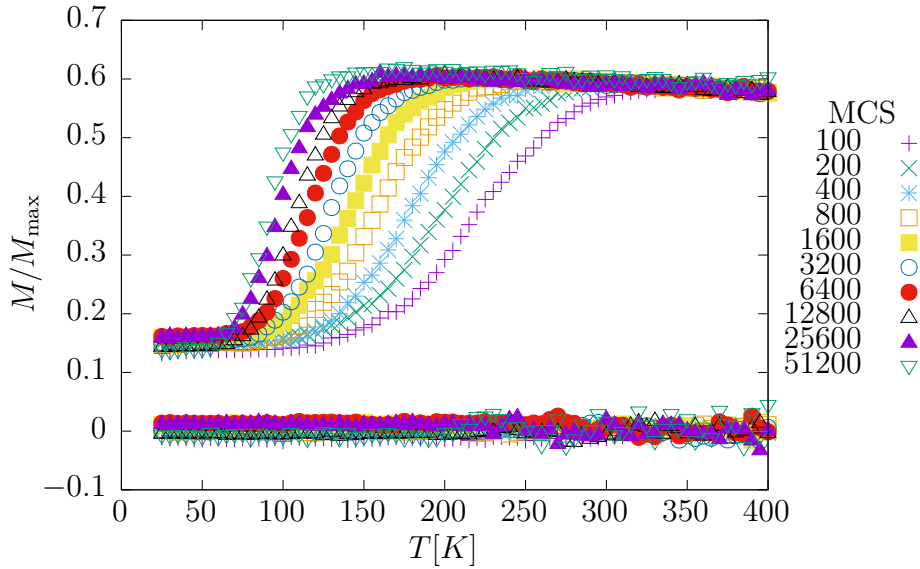


Figure 25: ZFC curves with varying number of Monte-Carlo steps (MCS)

In contrast to the previous case, the number of MCS at fixed test-vector length is a well-behaving parameter. An increase of MCS yields lower T_B which is the expected behaviour because any magnetic moment is allowed more time per temperature step to find its equilibrium. However, an increase of MCS leads to a convergent series of possible observations, precisely because our algorithm is based on Markov-Chains.

In contrast to the previous example, the parameter change here does not constitute a different physical system being studied. For higher MCS, we enhance the statistical quality of our simulation.

4.2.3.4 Applied Magnetic Field

Figure 26 shows simulations of the ZFC-curve for different applied magnetic fields after cooling in zero-field, but otherwise same parameters.

- number of magnetic moments: 1000
- Test vector modulus $|\mathbf{d}_m| = 1$
- Monte-Carlo steps per point: $2000^{(\text{relaxation})} + 10^{(\text{averaging})}$

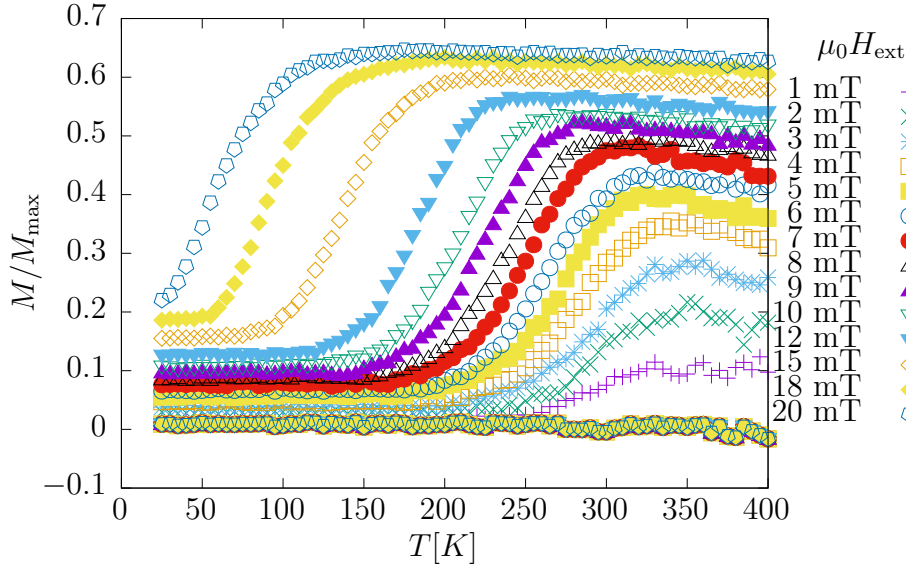


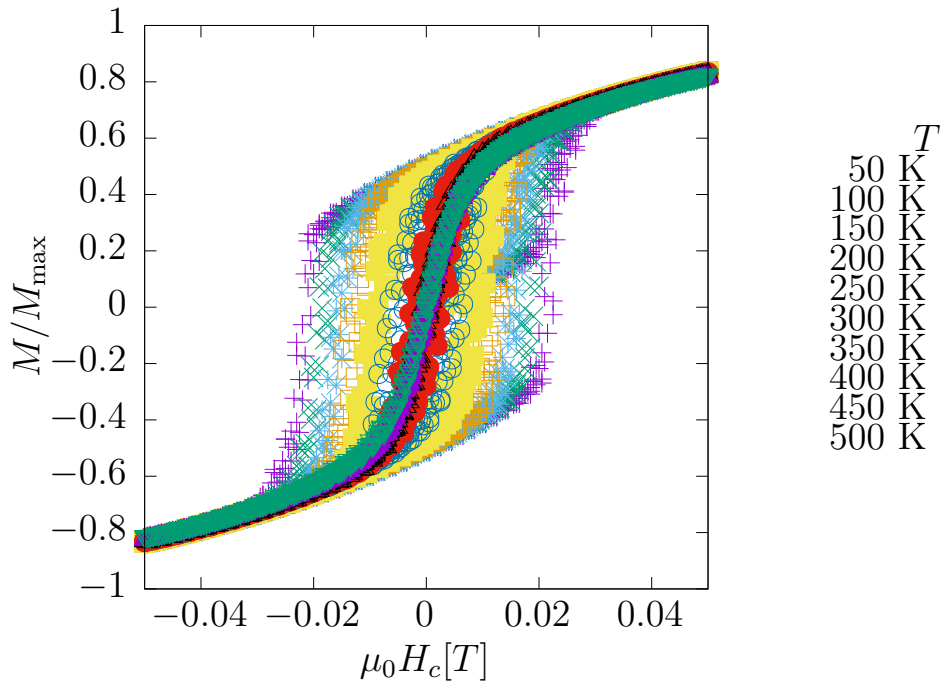
Figure 26: ZFC curves with varying applied field $\mu_0 H_{ext}$

The resulting series of ZFC curves shows, as expected, that T_B decreases when the applied field is increased.

4.2.4 Hysteresis Plots

Alternatively to cooling and heating the sample at discrete applied magnetic fields, it is insightful to apply varying external fields at constant temperature on the sample after it has been cooled down at zero external field.

In the limit of vanishing anisotropy constants, this procedure will reproduce our previous results for the isotropic paramagnet. For the more realistic case of finite KV , we will encounter the well-known phenomenon of open hystereses.



Comparing hysteresis loops at different temperatures will give a relationship between coercivity and temperature. The coercive field is defined as the strength of the applied magnetic field required to reduce the magnetization of the sample after it had been driven to saturation.

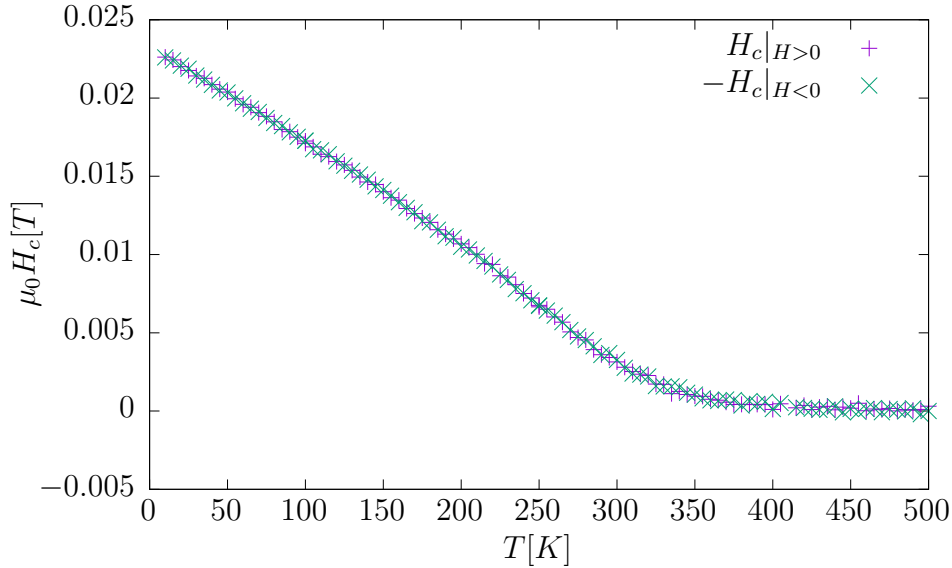


Figure 27: Temperature dependence of observed coercivity. The hysteresis is symmetric as shown by the overlapping of the curves

This behavior is also in accordance with experimental results. However, a theory of the precise analytical nature of the function $H_c = H_c(T)$ would have to involve the nature of modeled/implemented relaxation times τ as discussed in the context of Neél-Brown theory.

Another (technical) aspect is that the presented simulation results were achievable with far lesser computation time than comparable T-sweep simulations but with same statistical quality. Quite often it is observed that Metropolis-algorithm simulations at constant temperature yield much faster convergence.

Unfortunately, in the context of systems that feature frustration, such as most realistic magnetic systems do, experiments where the temperature is not held constant offer more insight.

4.2.5 ac-Susceptibility and Cole-Cole Plot

Finally, before moving on to interacting systems, we cover a third kind of sweep through parameter space. We want to investigate the AC-, or complex susceptibility χ_{ac} in the environment of a sinusoidal applied field at frequency ω

$$\chi_{ac}(\omega) = \chi'(\omega) - i\chi''(\omega) \quad \chi', \chi'' \in \mathbb{R}$$

Studying an explicitly time-dependent quantity is interesting from a technical point of view because we will directly make use of the interpretation of '1 Monte-Carlo step' as representative of a finite if extremely small period of time.

$$\mathbf{H}_{\text{ext}} \equiv \mathbf{H}(t_{MC}) = \mathbf{H}_0 \sin(\omega t_{MC})$$

is the physical field we want to simulate. We introduced a 'Monte-Carlo time' t_{MC} which we can implement in a straightforward fashion in a computer experiment by a certain number of Monte-Carlo steps. For χ', χ'' we then have

$$\begin{aligned} \chi'(\omega)\mathbf{H}_0 &= \frac{1}{N_{MC}} \sum_{t_{MC}=1}^{N_{MC}} \mathbf{M}(t_{MC}) \sin(\omega t_{MC}) \\ \chi''(\omega)\mathbf{H}_0 &= \frac{1}{N_{MC}} \sum_{t_{MC}=1}^{N_{MC}} \mathbf{M}(t_{MC}) \cos(\omega t_{MC}) \end{aligned}$$

Throughout this thesis we always consider magnetic samples that are considered linear and isotropic media. Thus, we can omit the vectors and χ is scalar instead of a tensor of degree ≥ 2 .

As one can plainly see from the definition above, high frequencies are implemented via a larger number of MC steps. This approach works fine if the computational effort behind one MCS is small, but it will be a great obstacle when treating interacting systems.

Figure 28 shows the results for the temperature dependence of the real and imaginary part of χ at fixed frequency.

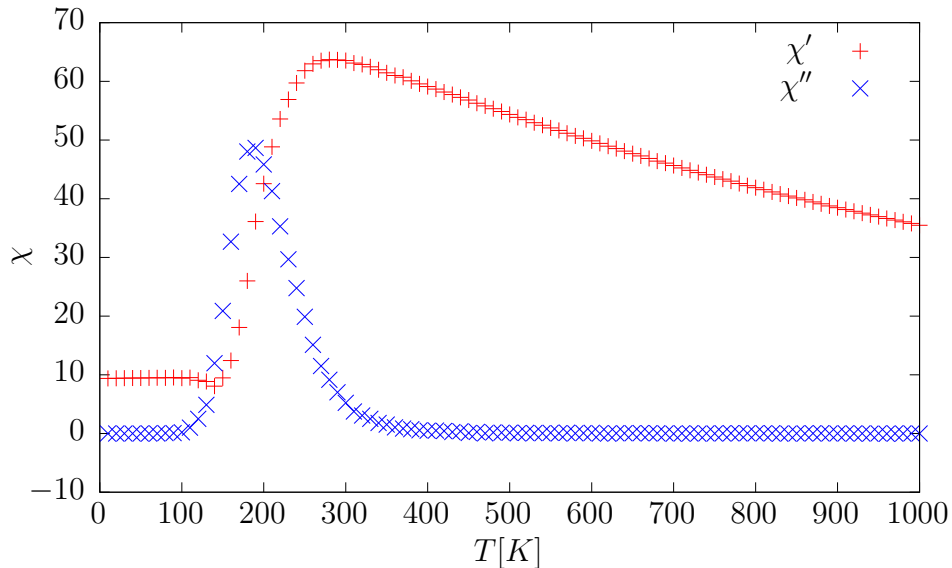


Figure 28: Real and imaginary part of the ac-susceptibility at $N_{\text{MCS}} = 10^6$

A frequency sweep at fixed temperature, conversely yields the Cole-Cole plot in Fig. 29:

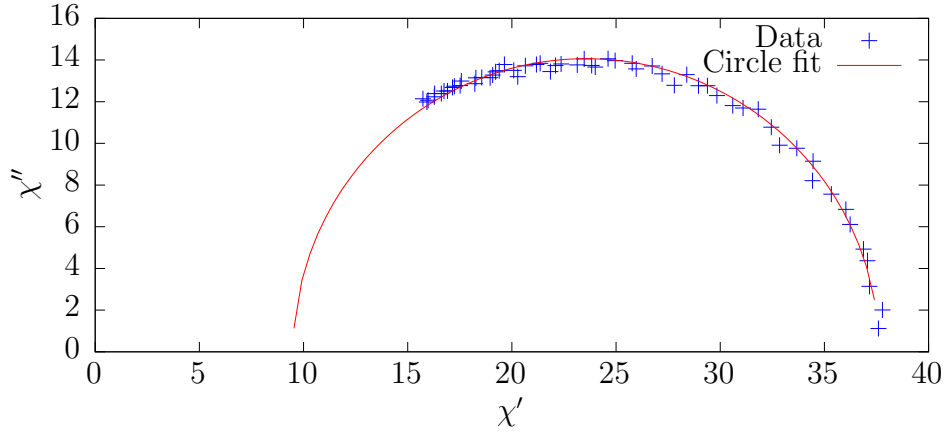


Figure 29: Cole-Cole plot and with fitting of a circle function from (4.9)

We find that our result confirms the expectation of a non-interacting system because a perfect semi-circle is observed. In contrast, an interacting system would show a strongly flattened circle.

4.3 Summary and Discussion of Results

Non-interacting systems have been comprehensively studied and we have collected sufficient evidence that our simulation approach is correctly implemented. We have found that simulation parameters such as the number of MCS per measurement have significant influence on any quantifiable observation.

5 Ferromagnetic Nanoparticles with Dipole-Dipole Interaction

From now on, we will consider nanoparticles forming supercrystals at realistic length scales (10^{-8} to 10^{-7} m). Here, it is no longer a realistic assumption to consider the nanoparticles as non-interacting. Instead, our model of supercrystals will include magnetic dipole-dipole interactions as additional energy contribution.

Theoretical aspects of this model are discussed first, especially computational challenges arising from any 2-particle interaction and the comparatively sparse theoretical knowledge regarding 3D models with a rather complicated interaction like the dipole-dipole interaction.

Motivated by this, we discuss widely used approximative approaches towards these models and re-introduce a method originally used in molecular physics in order to apply a customized version for our models.

5.1 Theory II

As noted, the only difference compared to the previous section is the introduction of realistic 2-particle interactions in our models. However, this small change has major implications for code efficiency and other aspects which were rather simple in the case of non-interacting nanoparticles.

5.1.1 Computational Aspects

Recalling the Metropolis-Hastings algorithm from (3.6) we see that an obvious bottleneck for code efficiency is the estimation of the total energy change of the system. This effect is far larger in case of 2-particle interactions because it will imply a $\mathcal{O}(N^2)$ -scaling with particle number, or correspondingly $\mathcal{O}(L^6)$ -scaling with edge length L .

On the other hand, we note that this energy estimation is entirely deterministic. This is important because we can then at least utilize multi-threaded computations to speed this step up. This will be done and discussed in the sections which do not employ different approximative theories. We note however that, purely from a computer efficiency point of view, it will always pay off to reduce the amount of 2-particle interaction computations.

5.1.2 Mermin-Wagner Theorem

Another challenge in this thesis is the absence of comprehensive theoretical knowledge what a dipolar 3D system is expected to behave like.

Although it is not the focus of this research, we recall an important result for sys-

tems of lower dimensionality, the Mermin-Wagner theorem [14]:

Mermin-Wagner Theorem

In 1D and 2D systems with sufficiently short-range interactions, no continuous symmetry can be broken spontaneously. This means that any thermal fluctuation offers sufficient perturbation destroying a possible ordering.

This result rigorously only applies to an isotropic Heisenberg ferromagnet, but generalizations towards various many-body systems exist [6]. This possesses rotational symmetry so that all the spin directions can be globally rotated without any additional energy cost. This means that long wavelength excitations, in which the spin state may deviate from its ground state value over a considerable distance, cost very little energy. Thus a fluctuation of the spins can be excited with very little energy cost. In one and two dimensions, they destroy the long range order. If, however, there is significant anisotropy there will be an energy cost associated with rotating the spins from their ground state value.

It turns out that the anisotropy energy penalty incurred by allowing these fluctuations increases with the square of R , the radius of the excitation, and hence the anisotropy energy will suppress all but the smallest of these non-linear fluctuations. It is the presence of such symmetry breaking fields which can stabilize long range order in two-dimensional systems. There is also a dipolar interaction between spins in real systems which, although much weaker than the exchange interaction, is anisotropic and can act in a similar way to suppress the growth of fluctuations.

5.1.3 Antiferromagnetic Part of the Dipole-Dipole Interaction

When writing down the full dipole-dipole energy between two magnetic moments $\mathbf{m}_1, \mathbf{m}_2$

$$\begin{aligned} E_{\text{dip}} &= \frac{\mu_0}{4\pi r^3} [\mathbf{m}_1 \cdot \mathbf{m}_2 - 3(\mathbf{m}_1 \cdot \hat{\mathbf{r}})(\mathbf{m}_2 \cdot \hat{\mathbf{r}})] \\ &= \underbrace{\frac{\mu_0}{4\pi r^3} (\mathbf{m}_1 \cdot \mathbf{m}_2)}_{=: E_{\text{afm}}} - \underbrace{\frac{3\mu_0}{4\pi r^3} (\mathbf{m}_1 \cdot \hat{\mathbf{r}})(\mathbf{m}_2 \cdot \hat{\mathbf{r}})}_{=: \tilde{E}_{\text{dip}}} \end{aligned} \quad (5.1)$$

we can identify two contributions which we call the *antiferromagnetic* part E_{afm} and residual

$$\tilde{E}_{\text{dip}} \equiv E_{\text{dip}} - E_{\text{afm}}$$

We can now imagine a hypothetical lattice model where the Hamilton function or Hamiltonian is completely defined by the 2-particle interaction given by $E_{\text{afm}} =$

$\frac{\mu_0}{4\pi r^3} (\mathbf{m}_1 \cdot \mathbf{m}_2)$. We observe that this resembles the conventional Heisenberg model Hamiltonian

$$\mathcal{H}_{\text{Heis}} = - \sum_{i,j} J_{ij} \mathbf{S}_1 \cdot \mathbf{S}_2 \quad (5.2)$$

where J_{ij} is the exchange integral between two spins $\mathbf{S}_1, \mathbf{S}_2$. The analogy we propose is therefore

$$\begin{aligned} E_{\text{afm}} &\leftrightarrow \mathcal{H}_{\text{Heis}} \\ \mathbf{m}_i &\leftrightarrow \mathbf{S}_i \\ \frac{\mu_0}{4\pi r^3} &\leftrightarrow -J_{ij} \end{aligned}$$

Most importantly, we see that our 'exchange integral'

$$-\frac{\mu_0}{4\pi r^3} \triangleq J_{ij} < 0 \quad \forall \text{ pairs } (i, j)$$

which implies that antiferromagnetism is the expected type of collective magnetism if the system has the appropriate translational symmetry.

5.1.4 Finite and Infinite Systems

It is possible to study finite systems non-approximatively. By this we mean that one can in principal evaluate the complete energy of any interacting system as long as the number of considered magnetic moments is finite. Computational constraints will however restrict such simulations in terms of maximum number of moments in the system:

As discussed in section 5.1.1, the computational effort necessary to exactly calculate the necessary energy terms for each MCS scales with $\mathcal{O}(N^2)$. The simulation of one measurement (i.e. one instance of the parameters T and $\mu_0 H_{\text{ext}}$) for a system of $N \sim 10^3$ particles may take as long as 1 hour on machines that are available to me. Realistically, any obtainable sample of self-assembled nanoparticles in a superlattice will consist of no fewer than $N \sim 10^9$ contributing superspins which will already imply an estimated computation time factor of 10^{12} . Therefore, what took 1 h before, would require several orders of magnitude longer.

This estimate ignores the fact that in the implementation used in this thesis (see appendix B.1.4) invariant parameters like site distances d and displacement vectors $\hat{\mathbf{r}}$ are calculated once and kept in working storage. For $N \sim 10^3$, this requires ~ 100 MB of RAM in the implementation given in the appendix. This could of course be cut down drastically if one sacrifices the flexibility of our code in terms of which superlattices can be put in, but it will still require $\sim 10^3$ KB of RAM and the same multiplication factor as before yields $\sim 10^9$ GB of required RAM. One could of

course perform simulations without putting these recurring parameters into storage, but this will in turn slow down the performance even further.

Because of these harsh limitations, we will focus on ensembles of $N < 10^5$ superspins. These are either considered

1. finite systems where surface effects will prohibit any meaningful insight into bulk behavior
2. supercells that are continued indefinitely with periodic boundary conditions in order to simulate bulk behavior

This thesis puts more emphasis on the second branch. Additional assumptions and approximations are required to effectively study interacting systems to a similar degree as was done for non-interacting systems in section 4.2.

5.1.5 Possible Approximations

We present a selection of possible approaches to simulate interacting lattice models effectively. We focus on methods that are still within the Metropolis scheme (3.6). This means that we focus on approximations that only affect the calculation of energies.

- **Approximations for finite systems**
 - Cut-off
- **Approximations for infinite systems with PBC**
 - Ewald method
 - Cut-off
 - Cut-off + Mean Field (Onsager)

The idea of introducing a cut-off length beyond which interactions are neglected is that the loss in precision of the calculated energy is far outweighed by the gain in computation efficiency.

5.1.6 Onsager Reaction Field Method

We will now cover in detail the adaptation of a mean-field theory applied mostly in molecular physics, for the purpose of getting reasonable energy estimations. It will offer a vast reduction of the needed computation time. A derivation from a general idea which implies a classical magnetostatic problem will be presented.

The mathematical problem itself is exactly solvable and will thus be presented thoroughly. Emerging parameters and their physical significance will be discussed. Finally, both the achieved accuracy in simulations and the performance from an efficiency standpoint will be discussed.

5.1.6.1 Motivation and Model

In molecular physics, research on electric dipolar particles in fluids is an important topic with a long history. With electric dipolar fluids it is important to develop approximate methods to estimate energies stemming from two-particle interaction such as the electric dipole energy, similar to our simulation studies.

The dipole-dipole interaction has the same form whether one talks about the energy of electric permanent dipoles $\mathbf{p}$ within an external electric field $\mathbf{E}$ or magnetic dipoles $\mathbf{m}$ in an applied $\mu_0\mathbf{H}$. Therefore, it appears reasonable to take previous work on approximative methods there as motivation for our work on magnetostatics.

Starting from the complete model with long-range interactions, the first step in the Onsager-Reaction-field method [16] is to introduce a cutoff radius R which limits the spatial extension of any electric field $\mathbf{E}_d$ produced by an electric dipole

$$\mathbf{E}_d(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \frac{1}{r^3} (3(\mathbf{p} \cdot \hat{\mathbf{r}})\hat{\mathbf{r}} - \mathbf{p}) \quad \mapsto \quad \mathbf{E}_d^{\text{ORF}}(\mathbf{r}) = \Theta(R - r) \mathbf{E}_d(\mathbf{r}) \quad (5.3)$$

with the Heaviside function Θ

$$\Theta(x) = \begin{cases} 0 & x < 0 \\ 1 & x \geq 0 \end{cases}$$

In the context of computer simulations, this is already the most important step of the entire approximation because it implies that only a fraction of interaction terms have to be computed with $\mathbf{E}_d^{\text{ORF}}$ instead of $\mathbf{E}_d$.

On the other hand, taking this new model-interaction on its own is obviously too naive because one would quickly come to the conclusion that there were no qualitative difference between bulk behavior or the behavior of a tiny set of interacting dipoles.

The 2nd part of the ORF-method therefore provides an idea how to model the effect of the environment outside the cut-off sphere on the dipole in the center. The idea is to model these magnetic moments as a continuous, linear, isotropic, polarized medium and imagine it reacting towards a superdipole which sits at the exact center of the sphere which otherwise is comprised of vacuum. This medium will have a relative permittivity³ $\epsilon_r > 1$ which is a real scalar and is a quantity which can also be calculated when measuring the total polarization of the entire set of electric

³More generally, $\epsilon_r \neq 1$ would suffice, but metamaterials with negative susceptibility $\chi_e \equiv \epsilon_r - 1$ are rarely considered

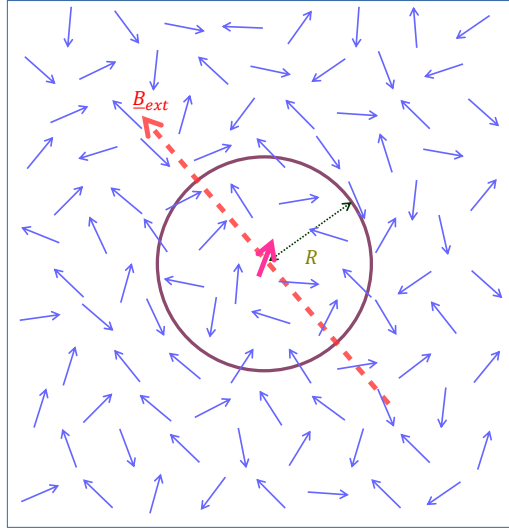


Figure 30: First step of the ORF method: Introduce cut-off R beyond which dipole-dipole interactions are not considered

dipoles in an external electric field.

From there, classical electrostatics will give an expectation for the *reaction field* $\mathbf{E}_{\text{RF}}$, an additional electric field which is the response of the surrounding medium towards the presence of the dipole.

As stated, the magnetic field $\mathbf{B}_d$ of a magnetic dipole has the same form as in the electric case, which is why we can just propose the following $\mathbf{B}_d^{\text{ORF}}$ providing a cut-off for the magnetic field analogously to (5.3)

$$\mathbf{B}_d(\mathbf{r}) = \frac{\mu_0}{4\pi} \frac{1}{r^3} (3(\mathbf{m} \cdot \hat{\mathbf{r}})\hat{\mathbf{r}} - \mathbf{m}) \quad \mapsto \quad \mathbf{B}_d^{\text{ORF}}(\mathbf{r}) = \Theta(R - r) \mathbf{B}_d(\mathbf{r}) \quad (5.4)$$

As in the original case of electric dipolar molecules, we now have to make an estimate for any environment of the magnetic dipole beyond the cut-off radius R : We imagine a continuous, linear, isotropic, magnetized medium with the relative permeability $\mu_r > 1$ surrounding the cut-off sphere which is comprised of vacuum ($\mu_r = 1$) and a magnetic super-dipole at its center. This super-dipole again is the direct sum of all magnetic moments contained in the cut-off sphere. The model is illustrated in figures 30 and 31.

5.1.6.2 Solution of the Magnetostatic Problem

Vector calculus states that any curl-free vector field is the gradient of a scalar field⁴.

⁴Provided that mathematical conditions like continuity, differentiability and correct asymptotic behavior for $r \rightarrow \infty$ are met.

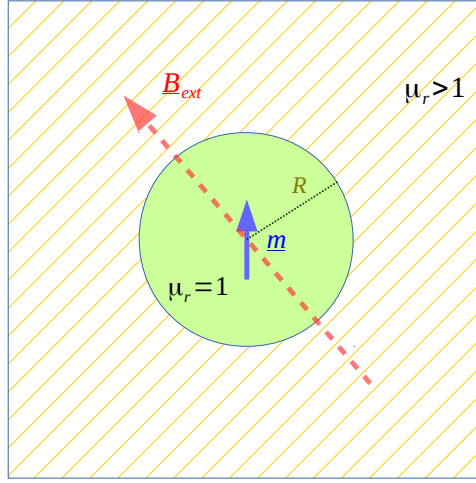


Figure 31: Second step of the ORF method: Onsager Reaction Field model as a problem in classical magnetostatics

This means

$$\nabla \times \mathbf{E} = 0 \quad \Rightarrow \quad \exists \phi(\mathbf{r}) : -\nabla \phi = \mathbf{E} \quad (5.5)$$

Therefore, the solution of boundary problems for any stationary *electric* field $\mathbf{E}$ and its potential ϕ correspond to a given charge density $\rho_e = \rho_e(\mathbf{r})$ via

$$\begin{aligned} \nabla \phi &= -\mathbf{E} \\ \text{div } \mathbf{E} &= \frac{\rho_e}{\epsilon_0} \\ \Rightarrow \quad \Delta \phi &= -\frac{\rho_e}{\epsilon_0} \end{aligned} \quad (5.6)$$

which is the well-known Poisson equation. Any $\phi(r)$ that solves the Poisson equation and also satisfies boundary conditions given by materials etc. is the unique solution to such a problem.

Finding a solution to (5.6) is relatively easy if the problem has any symmetries. Because of the Maxwell equation $\nabla \cdot \mathbf{B} = 0$ there is no analogous scalar potential for magnetic fields. Instead, one has a not easily found *vector* potential $\mathbf{A}$ such that $\mathbf{B} = \nabla \times \mathbf{A}$. This is given via the Biot-Savart law

$$\mathbf{B}(\mathbf{r}) = \frac{\mu_0}{4\pi} \int_V \frac{\mathbf{j} \, dV \times \mathbf{r}'}{|\mathbf{r}'|^3}$$

but cannot be evaluated easily since we first would have to find the correct static current density $\mathbf{j}$ which represents our permanent magnetic dipole $\mathbf{m}$.

It is therefore not immediately obvious that the derivation of an appropriate reaction field $\mathbf{B}_{RF}$ will be as easy as in the dielectric case.

However, we realize that there is no *free* current density in our problem, only *bound* currents which induce magnetization, i.e. a dipole-moment density. This will facilitate the mathematics immensely in the following way:

We use the auxiliary field $\mathbf{H}$ and a decomposition of the total charge current $\mathbf{j}$ into bound and free part:

$$\mathbf{H} = \frac{1}{\mu_0} \mathbf{B} - \mathbf{M} \quad (5.7)$$

$$\mathbf{j} = \mathbf{j}_{\text{free}} + \mathbf{j}_{\text{bound}} \quad (5.8)$$

$$\mathbf{j}_{\text{bound}} = \nabla \times \mathbf{M} \quad (5.9)$$

We study the derivatives of H :

$$\nabla \cdot \mathbf{H} = \frac{1}{\mu_0} \nabla \cdot \mathbf{B} - \nabla \cdot \mathbf{M} = -\nabla \cdot \mathbf{M} \quad (5.10)$$

$$\nabla \times \mathbf{H} = \frac{1}{\mu_0} \nabla \times \mathbf{B} - \nabla \times \mathbf{M} = \mathbf{j} - \mathbf{j}_{\text{bound}} = \mathbf{j}_{\text{free}}$$

Since there is no free current, we have found analogously to (5.5)

$$\nabla \times \mathbf{H} = 0 \quad \Rightarrow \quad \exists W(\mathbf{r}) : -\nabla W = \mathbf{H} \quad (5.11)$$

Combining (5.11) and (5.10) we have found the

Poisson equation of the magnetic potential

$$\Delta W = \nabla \cdot \mathbf{M} \quad (5.12)$$

just like (5.6) for electric fields. Now we can also make explicit use of our assumption that the outside medium is linear and isotropic:

$$\begin{aligned} \mathbf{M}_i &\stackrel{\text{linear}}{=} \sum_{j=1}^3 \chi_{ij}^{(m)} \mathbf{H}_j + O(\mathbf{H}^2) \\ \chi_{ij}^{(m)} &\stackrel{\text{isotropic}}{=} \chi_m \delta_{ij} \\ \Rightarrow \quad \mathbf{M} &= \chi_m \mathbf{H} \end{aligned}$$

Most importantly for our calculations below, this implies that $\mathbf{B}$ and $\mathbf{H}$ are related to each other simply by

$$\mathbf{B} \equiv \mu_0(\mathbf{H} + \mathbf{M}) = \mu_0\mathbf{H} + \mu_0\chi_m\mathbf{H} = \mu\mathbf{H} \quad (5.13)$$

where

$$\mu = \mu_0(1 + \chi_m) \equiv \mu_0 \mu_r \quad (5.14)$$

If we assume that the surrounding medium with magnetization M is 'roughly paramagnetic'

$$1 < \mu_r \lesssim 100$$

this assumption seems reasonable.

Very importantly, we hereby have a $\mathbf{H}_{ext}$ which is *not* constant throughout $\mathbb{R}^3$ like $\mathbf{B}_{ext}$ but piecewise defined as

$$\mathbf{H}_{ext} = \mathbf{B}_{ext} \begin{cases} \frac{1}{\mu_0} & r < R \\ \frac{1}{\mu_0 \mu_r} & r > R \end{cases} \quad (5.15)$$

In this very special case of magnetostatic problems, the boundary conditions given by the Maxwell equations look completely analogous to the electrostatic case via

$$W \leftrightarrow \phi$$

$$\mathbf{H} \leftrightarrow \mathbf{E}$$

$$\mu_r \leftrightarrow \epsilon_r$$

Finally, we arrive at the following set of differential equations and boundary conditions.

$$\textbf{Boundary conditions for the magnetic Poisson equation} \quad (5.16)$$

$$W(r \rightarrow 0) \rightarrow \frac{\mathbf{m} \cdot \mathbf{r}}{4\pi r^3}$$

$$W(r \rightarrow \infty) \rightarrow -\mathbf{H}_{ext} \cdot \mathbf{r}$$

$$W(r = R) = \text{continuous}$$

$$\mathbf{H}_{in}^\perp = \mu_r \mathbf{H}_{out}^\perp$$

$$\mathbf{H}_{in}^\parallel = \mathbf{H}_{out}^\parallel$$

Again, note that $\mathbf{m}$ now represents the total sum of dipole moments within V_c :

$$\mathbf{m} \equiv \sum_{V_c} \mathbf{m}_i \quad (5.17)$$

With (5.16), the problem is unfortunately very unsymmetric because the angle between $\mathbf{m}$ and $\mathbf{H}_{ext}$ will take arbitrary values.

In order to be able to use techniques for problems with azimuthal boundary conditions, we split the problem in two parts where either the cumulated magnetic moment $\mathbf{m}$ or the external field $\mathbf{B}_{ext} = \mu \mathbf{H}_{ext}$ are a symmetry axis:

$$\begin{aligned} W^I(r \rightarrow 0) &\rightarrow \frac{\mathbf{m} \cdot \mathbf{r}}{4\pi r^3} \\ W^I(r \rightarrow \infty) &\rightarrow 0 \\ W^I(r = R) &= \text{continuous} \\ \mathbf{H}_{in}^{I,\perp} &= \mu_r \mathbf{H}_{out}^{I,\perp} \\ \mathbf{H}_{in}^{I,\parallel} &= \mathbf{H}_{out}^{I,\parallel} \end{aligned}$$

$$\begin{aligned} W^{II}(r \rightarrow 0) &\rightarrow 0 \\ W^{II}(r \rightarrow \infty) &\rightarrow -\mathbf{H}_{ext} \cdot \mathbf{r} \\ W^{II}(r = R) &= \text{continuous} \\ \mathbf{H}_{in}^{II,\perp} &= \mu_r \mathbf{H}_{out}^{II,\perp} \\ \mathbf{H}_{in}^{II,\parallel} &= \mathbf{H}_{out}^{II,\parallel} \end{aligned}$$

The explicit derivation makes use of techniques to solve the homogenous Laplace equation

$$\nabla^2 W = 0$$

and is given in the appendix A.1.

The coordinate-free representation for the solution of the two problems is:

$$\begin{aligned} W_{in}^I(\mathbf{r}) &= \frac{\mathbf{m} \cdot \mathbf{r}}{4\pi} \left(\frac{1}{r^3} + \frac{1}{R^3} \frac{1 - \mu_r}{1 + 2\mu_r} \right) \\ W_{out}^I(\mathbf{r}) &= \frac{\mathbf{m} \cdot \mathbf{r}}{4\pi r^3} \frac{3}{1 + 2\mu_r} \\ W_{in}^{II}(\mathbf{r}) &= -\mathbf{H}_{ext} \cdot \mathbf{r} \frac{3\mu_r}{1 + 2\mu_r} \\ W_{out}^{II}(\mathbf{r}) &= \mathbf{H}_{ext} \cdot \mathbf{r} \left(\frac{R^3}{r^3} \frac{1 - \mu_r}{1 + 2\mu_r} - 1 \right) \end{aligned}$$

and direct summation yields the solution of the original problem:

$$W(\mathbf{r}) = \begin{cases} \mathbf{m} \cdot \mathbf{r} \frac{1}{4\pi} \left(\frac{1}{r^3} + \frac{1}{R^3} \frac{1-\mu_r}{1+2\mu_r} \right) - \mathbf{H}_{ext} \cdot \mathbf{r} \frac{3\mu_r}{1+2\mu_r} & r < R \\ \mathbf{m} \cdot \mathbf{r} \frac{1}{4\pi r^3} - \frac{3}{1+2\mu_r} + \mathbf{H}_{ext} \cdot \mathbf{r} \left(\frac{R^3}{r^3} \frac{1-\mu_r}{1+2\mu_r} - 1 \right) & r > R \end{cases} \quad (5.18)$$

We can now identify the so-called reaction field that has a finite value at $r = 0$:

$$\mathbf{H}_{RF} = - \left(\mathbf{m} \frac{1}{2\pi R^3} \frac{1-\mu_r}{1+2\mu_r} - \mathbf{H}_{ext} \frac{3\mu_r}{1+2\mu_r} \right) \quad (5.19)$$

Introducing μ_r -depending constants

$$\gamma_{\mathbf{m}} := \frac{1-\mu_r}{1+2\mu_r} \quad \gamma_{\mathbf{h}} := -\frac{3\mu_r}{1+2\mu_r} \quad (5.20)$$

our estimate for the energy of one superspin $\mathbf{s}$ due to the external field and the dipolar environment is therefore:

$$\begin{aligned} E_{RF} &= -\mathbf{s} \cdot \mathbf{B}_{RF} = -\mu_0 \mathbf{s} \cdot \mathbf{H}_{RF} \\ E_{RF} &= \mathbf{s} \cdot \left(\mathbf{m} \frac{\mu_0}{2\pi R^3} \gamma_{\mathbf{m}} + \mathbf{H}_{ext} \mu_0 \gamma_{\mathbf{h}} \right) \\ E_{RF} &= \mathbf{s} \cdot \left(\mathbf{m} \frac{\mu_0}{2\pi R^3} \frac{1-\mu_r}{1+2\mu_r} - \mathbf{B}_{ext} \frac{3\mu_r}{1+2\mu_r} \right) \end{aligned} \quad (5.21)$$

Both the $\mathbf{m}$ - and $\mathbf{B}_{ext}$ term have properties that should be discussed further.

We calculate μ_r via

$$\mu_r = 1 + \chi_m = 1 + \frac{M}{H_{ext}} = 1 + \frac{\mu_0 M}{B_{ext}} \quad (5.22)$$

where $M = |\mathbf{M}|$ is the magnetization, i.e. the total magnetic dipole density which is assumed to be linearly dependent of $B_{ext} = |\mathbf{B}_{ext}|$ throughout our approximation.

5.1.6.3 Modelling parameters

As a general rule, the reaction field encourages configurations within the cut-off sphere with large cumulative magnetic moment via a parallel auxiliary field because

$$-1/2 < \frac{1-\mu_r}{1+2\mu_r} < 0 \quad \mu_r > 1 \quad (5.23)$$

This term therefore indeed deserves the name 'reaction field' because this auxiliary field is the response of the imagined medium towards any net magnetic moment in the sphere.

The second term means that applied magnetic field acting on any single magnetic

moment is amplified because of the surrounding medium since

$$-3/2 < \frac{-3\mu_r}{1+2\mu_r} < -1 \quad \mu_r > 1 \quad (5.24)$$

At first look, this seems like the Onsager approach leads to inconsistencies in the limit of non-interacting systems. This is ultimately not the case, as shown in 5.1.6.6.

5.1.6.4 Cut-off Radius

The radius R largely determines the computational effort that an energy estimate with this approximation still requires. Intuitively one would expect that a smaller cutoff radius implies an increase of the necessary correction, which is indeed the case. The fact that the correction is inversely proportional to the volume of the cut-off sphere means that it is also inversely proportional to the number of superspins that are treated exactly within our energy-estimation scheme which seems reasonable.

Because we are always dealing with a discrete dipole distribution, we must however specify how R is implemented. There will always be an interval of possible R values whose corresponding cut-off spheres would contain the same magnetic moments. Without additional criteria, R could thus be chosen arbitrary which would however limit comparability between simulations.

We found that a consistent and reasonable estimate would be choosing *the greatest possible R that does not span additional sites outside the cut-off sphere*. This implies that no internal site touches the boundary of the sphere as seen in figure 32, and the reaction field takes on the lowest possible absolute value. In any case, R has to

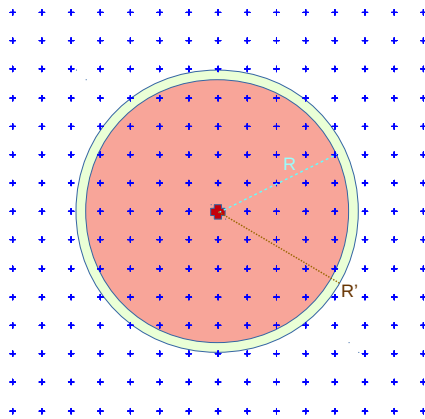


Figure 32: Choice of R for reaction field estimation. Blue crosses signify magnetic sites. Both the cyan and salmon colored circles contain the exact same sites. They represent the smallest and biggest possible choices for R

be chosen consistently in order to have comparable simulation results since e.g. a 10% increase of R already means that the $\mathbf{m}$ -factor is cut in half.

Simulations were mostly done on *fcc*-super lattices which have a very high degree of spatial symmetry so that the computation of the best choice for R needs to be carried out only once for any choice of 'modelling' cut-off.

5.1.6.5 Determination of the relative permeability

The recipe (5.22) given above has the advantage of being a self-consistent procedure because each magnetic moment will be updated regularly during the simulation of one measurement point and all under the same conditions. This means that the chosen μ_r indeed becomes the best choice self-fulfillingly.

This however holds only true if the global susceptibility remains roughly constant throughout these measurements. This is obviously not true e.g. during the process of a ZFC-FC curve where the applied magnetic field has only one value, yet the magnetization is dependent on the recent history and temperature of the sample.

The hitherto implemented scheme thus suffers at least from temporal retardation, which becomes an issue if the goal of the simulation is to pin down an exact value for the blocking temperature.

Even worse, in the absence of applied fields the recipe (5.22) becomes ill-defined and one has to either retroactively impose values from e.g. high-temperature values in the ZFC-curve or impose a theoretical expectation like the Langevin value for paramagnets

$$\mu_r^{\text{Langevin}} = 1 + \chi^{\text{Langevin}} = 1 + n\mu_0 \frac{(M_s V)^2}{3k_B T}$$

which we only used as a very first guess. In subsequent simulations, the value was indeed changed to be the value found for the tail-end of ZFC curves which let our μ_r converge towards $\mu_r \sim 15$ in zero-field.

The exact value of μ_r would of course be an interesting property/result in itself, however its influence of our reaction field is limited. The two prefactors in (5.20) are shown in figure 33.

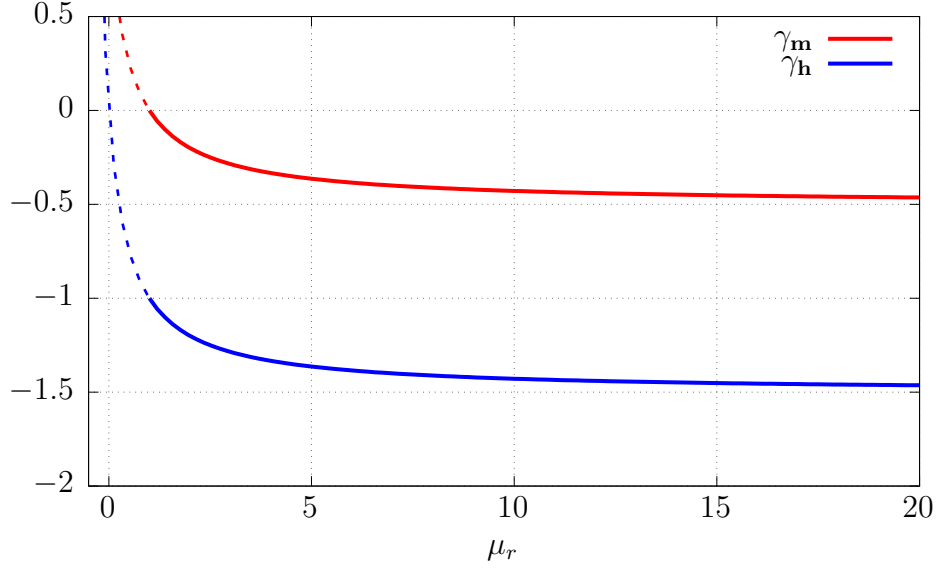


Figure 33: Dependence of correction factors on relative permeability. Only $\mu_r > 1$ (solid) applies in our simulations

The prefactors vary only very slightly for $\mu_r \gtrsim 10$, and the applied-field correction term does not even apply in zero-field. Therefore, we can at least have confidence that the precise value of our assumed μ_r is of less importance than e.g. the validity of the assumptions regarding the material in (5.13)

5.1.6.6 Consistency with non-interacting limit

It is expected that the influence of the dipole-dipole interaction on the sample diminishes quickly with an increase of the lattice constant of the supercrystal a_0 . This will be reflected automatically for the interactions that are considered in an exact matter within V_c . The $\mathbf{m}$ -factor in the reaction-field term also reflects an increase of the lattice constant automatically because R per design scales automatically with any change of the lattice constant.

The $\mathbf{B}_{ext}$ -factor does not scale directly with the spatial dimensions of the sample. The scale of the system is instead only influential via μ_r because the magnetization M in (5.22) is the dipole moment *density* which means that if only the lattice constant increases without adding further magnetic dipoles, we have for μ_r , γ_m , γ_h

$$\left. \begin{array}{ll} \mu_r & \rightarrow 1 \\ \gamma_m = \frac{1-\mu_r}{1+2\mu_r} & \rightarrow 0 \\ \gamma_h = \frac{3\mu_r}{1+2\mu_r} & \rightarrow 1 \end{array} \right\} \quad \text{for } a_0 \rightarrow \infty$$

This means that the $\mathbf{m}$ -term vanishes even faster than $\propto 1/R^3$.

$\gamma_h \rightarrow 1$ means that there is no amplification via the magnetic medium anymore, which is also to be expected.

In total, we arrive for large lattice constants at a vanishing reaction field so that

only the superspins within V_c will interact via dipole-dipole interaction and the unaltered externally applied field. As shown in figure 34, we get a smooth transition of the ZFC/FC curves towards the non-interacting case as soon as $\frac{a}{a_0} \sim 5$ where a_0 is chosen such that for $a \leq a_0$ the behavior is significantly different due to 2-particle interactions: $a_0 \simeq 25$ nm.

This means that the behavior of our approximative theory is largely correct in the non-interacting limit.

To understand what happens at $a = a_0$, we calculate the dipole-dipole energy of one spin in the environment of ≥ 100 randomly oriented neighbouring spins. This component of the total potential that is independent from all anisotropy axes is given in figure 35.

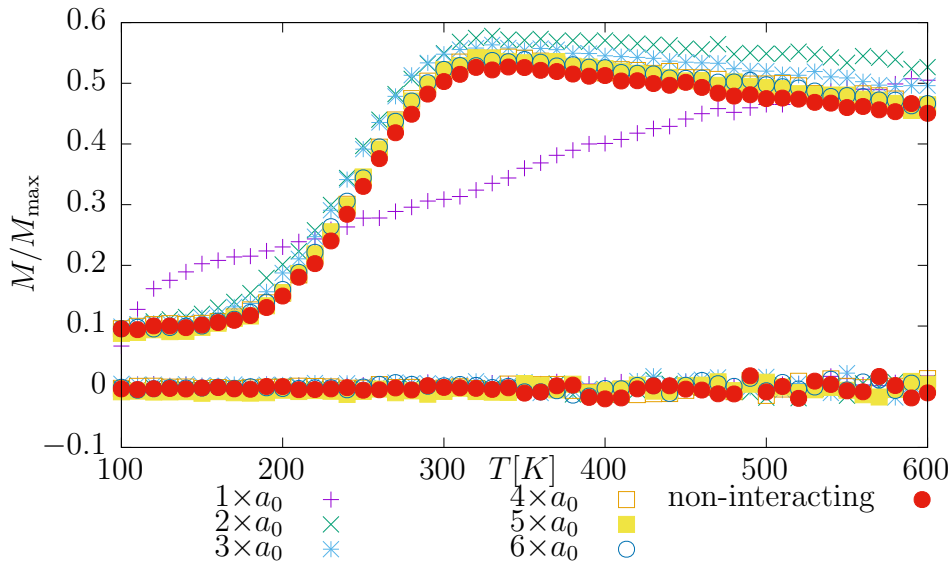


Figure 34: Convergence of ORF simulations of the ZFC curve towards the non-interacting limit

The relevant information of this is that there is always exactly one global minimum produced by the dipole fields. Even local minima are only rarely observed. If one adds this potential to the known Stoner-Wohlfarth results in 4.1.3, one observes in most cases that additionally to a change in position of the entire potential landscape, that the potential wall between the two local minima increases. Since the blocking temperature is predominantly associated with this potential wall, one can already assume that the dipole-dipole interaction will increase the blocking temperature.

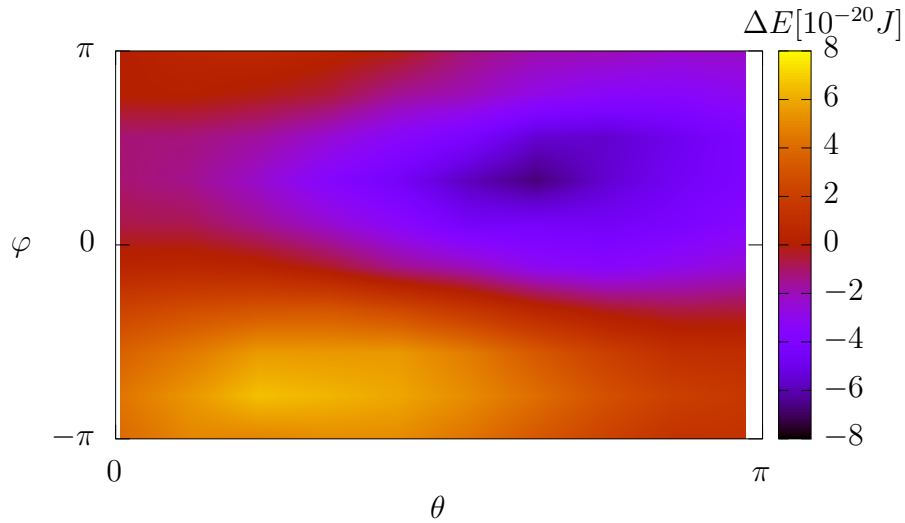


Figure 35: Potential landscape for a superspin in a 8x8x8 magnetic supercell (fcc). Plotted are energy differences compared to the energy for orientation $\theta = \pi/2$, $\varphi = 0$. Any cut-off of more than 30 contributing neighbours leads to an image that is indistinguishable at this resolution

5.2 Simulation Results

We now present simulation results obtained by studying systems with interacting nanoparticles.

5.2.1 Groundstates in Periodic, Dipolar 3D Systems

We discuss a topic that is motivated more from pure statistical physics point of view, than in the context of self-assembled magnetic nanoparticles. We consider 3D crystal systems whose total energy is exclusively determined by the magnetic dipole-dipole interaction of magnetic moments positioned on the lattice sites. The aim is an investigation of a purely dipolar ground state in certain lattice structures.

5.2.1.1 Results in the Limit of Vanishing Magnetocrystalline Anisotropy

We realize that the question above is equivalent to the physically not viable case of self-assembled magnetic nanoparticles in the limit of vanishing magnetocrystalline anisotropy at each lattice site. Simulations can be therefore carried out in a straightforward fashion with our existing methods. We consider systems that are initiated at very high temperatures and then cooled down with zero applied field. We pay particular attention to the energy of the system and discuss the superspin landscape at $T \rightarrow 0$.

With the help of the self-developed program which is documented in B.2, we will produce figures like figure 36. In all of these figures, all spins are grouped into

sublattices that in most cases span the magnetic super cell and require all of their member sites to be parallel aligned within a certain margin of error.

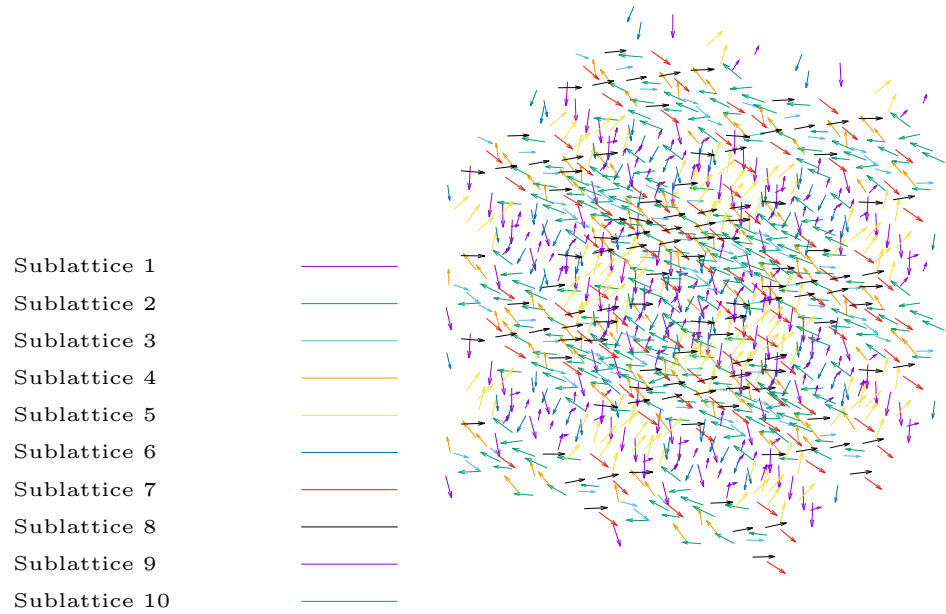


Figure 36: 8x8x8 magnetic super cell (*fcc*) at $T = 1$. Spin distribution of one start configuration

In this thesis, I mostly show instances where these sublattices have the peculiar property of being essentially 2-dimensional so that one can find a perspective that makes the spin structure readable by a projection on a plane, as seen in figure 37.

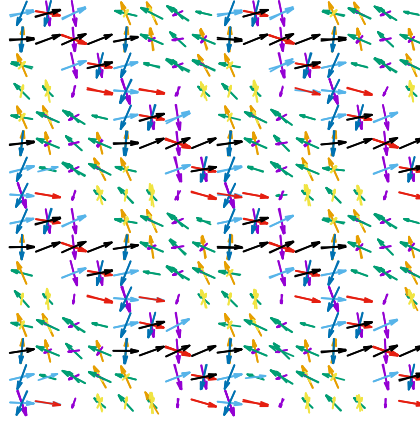


Figure 37: Rotated view of figure 36

5.2.1.2 Series of 'Groundstates' Depending on Different Parameters in the Onsager Approximation

We present several low energy states and their spin structure that were obtained with varying spin initializations. Although different cut-off radii were employed to further increase the variety of simulated configurations, the following selection of low-energy structures were found independently from these respective choices.

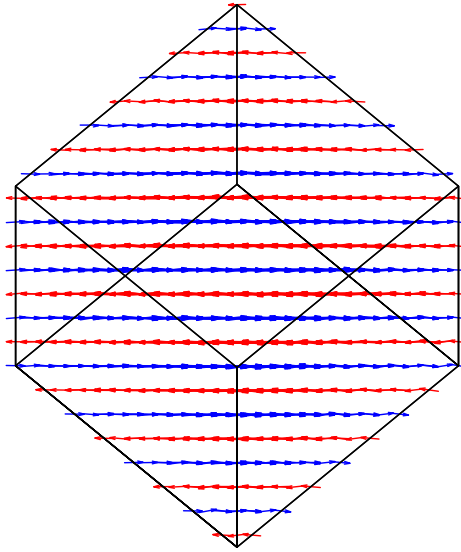


Figure 38: 2 sublattices, inspecific view

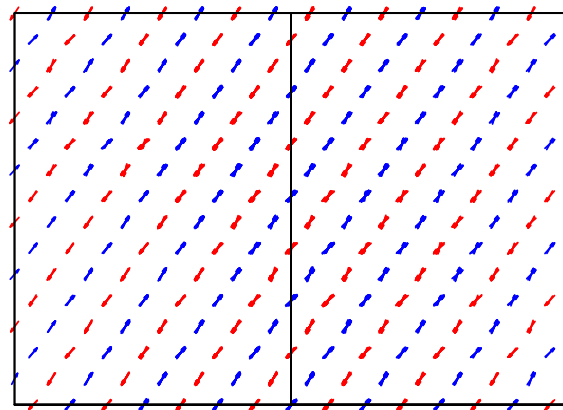


Figure 39: Rotated view of figure 38

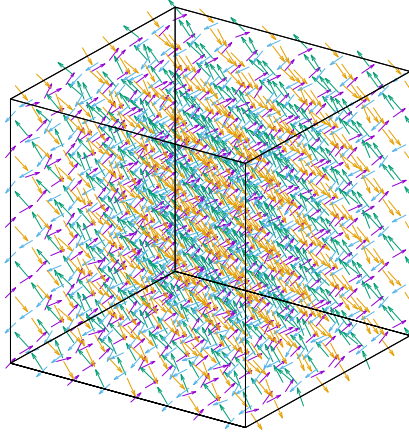


Figure 40: 4 sublattices, inspecific view

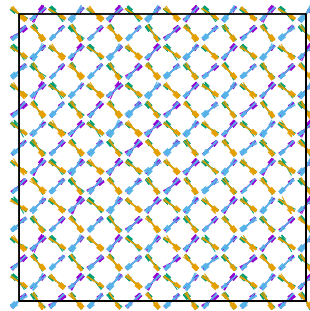


Figure 41: Rotated view of figure 40

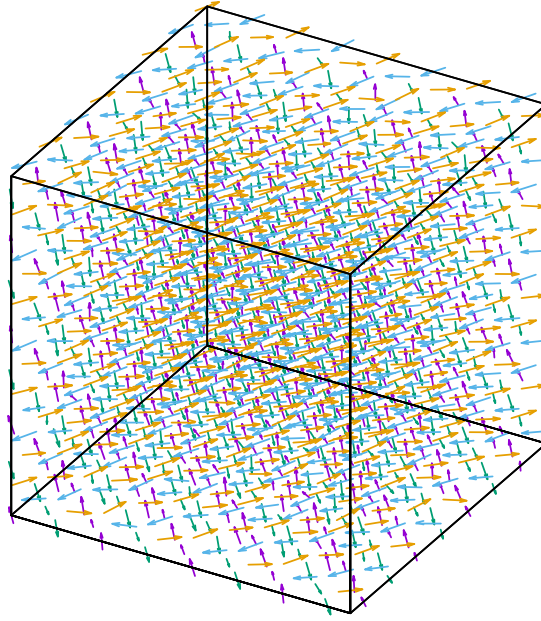


Figure 42: 4 sublattices, inspecific view

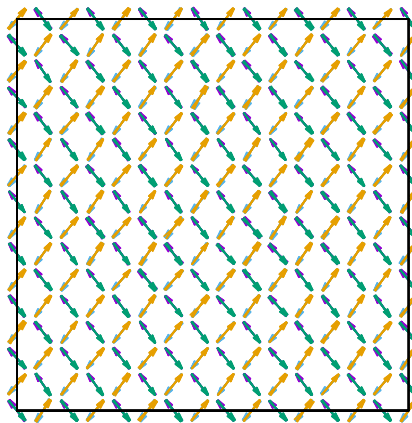


Figure 43: Rotated view of figure 42

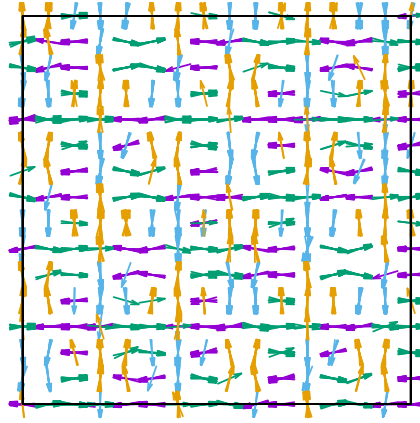


Figure 44: 4-6 sublattices as a more complex configuration, rotated view

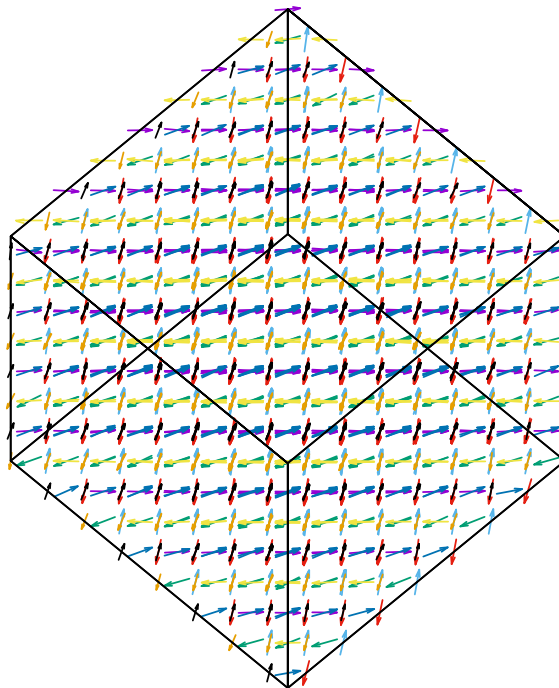


Figure 45: 8 sublattices, inspecific view

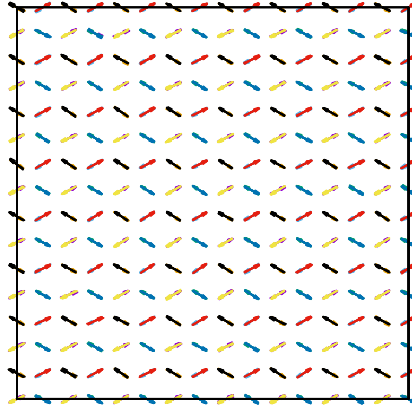


Figure 46: Rotated view of figure 45, yz-plane

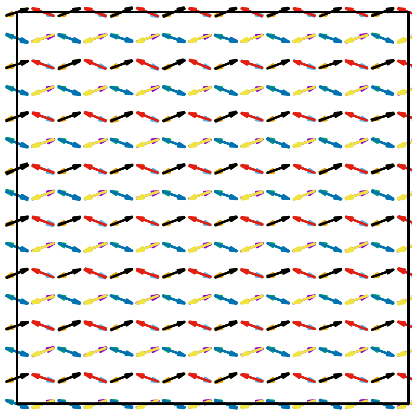


Figure 47: Rotated view of figure 45, xz-plane

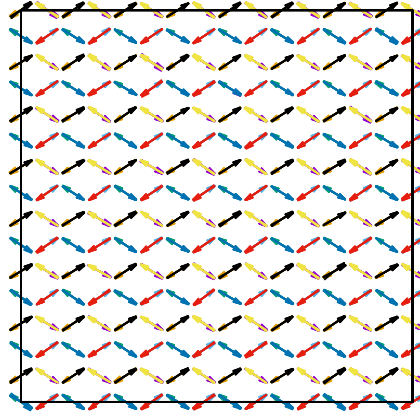


Figure 48: Rotated view of figure 45, xy-plane

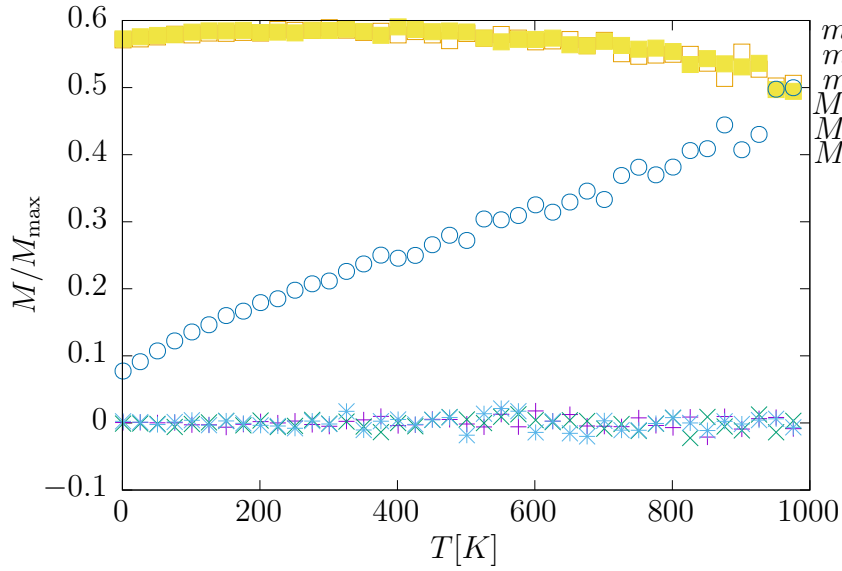


Figure 49: Temperature dependence of the magnetization components for the sample as shown in both figures 42 and 43. M_x , M_y , M_z are defined in (5.25). By these 'components', the 2-dimensional character of the low-energy state corresponds to the vanishing of 'MZ'

It is useful to define the following quantities:

$$M\alpha := \sum_{i=1}^N |m_{\alpha}^i| \quad \alpha = x, y, z \quad (5.25)$$

This means that e.g. $Mx = 0$ if the x -component of the magnetization of each site is zero and *not* just randomly distributed in $(-1, 1)$. Therefore, Mx measures if the x -component of the magnetization vanishes completely, and the system therefore does not have a 3-dimensional, but a layered 2-dimensional magnetization landscape.

Apart from this often occurring reduction of dimensionality, it has always been observed that an even number of antiparallel sublattices emerges at low temperatures. However, we cannot call this behaviour antiferromagnetic because no spontaneous symmetry breaking is observed. Conversely, this 'phase transition' happens steadily but slowly in an arbitrarily large temperature region.

5.2.1.3 Reconsidering Non-Vanishing Anisotropy

We connect the previous chapter back to the more realistic setting of magnetic nanoparticles such as maghemite nanoparticles. We discuss how stable the findings of chapter 5.2.1.2 are with regards to non-zero anisotropy energies.

In short, employing realistic parameters of the maghemite nanoparticles in order to study the number and character of sublattices at low temperatures yields a clear but disappointing result: No long range order can be detected, meaning that the random anisotropy changes the local potential landscapes too strongly.

On a more positive note, we can therefore study ZFC curves quite well with the help of the ORF method because it is even more viable if there is no (short-range) magnetic order at the temperatures we consider. Simulation results are given in figures 50 and 51. Because of the more complicated potential landscape, it is necessary to make the simulations with a smaller test-vector length than in the non-interacting case. Otherwise, local potential minima maybe insufficiently scanned by our algorithm and we would find non-physical dynamics.

Both simulations are performed with $d_m = 0.1 \ll 1$, but the first plot shows that one must then also increase the number of Monte-Carlo steps per measurement: It has only been increased to 2000 from 1000 in the non-interacting examples earlier. The different shape and unrealistically high blocking temperature show that this simulation does not represent nature, and one has to increase the number of MCS even further. Of course, the real computation time also increases linearly.

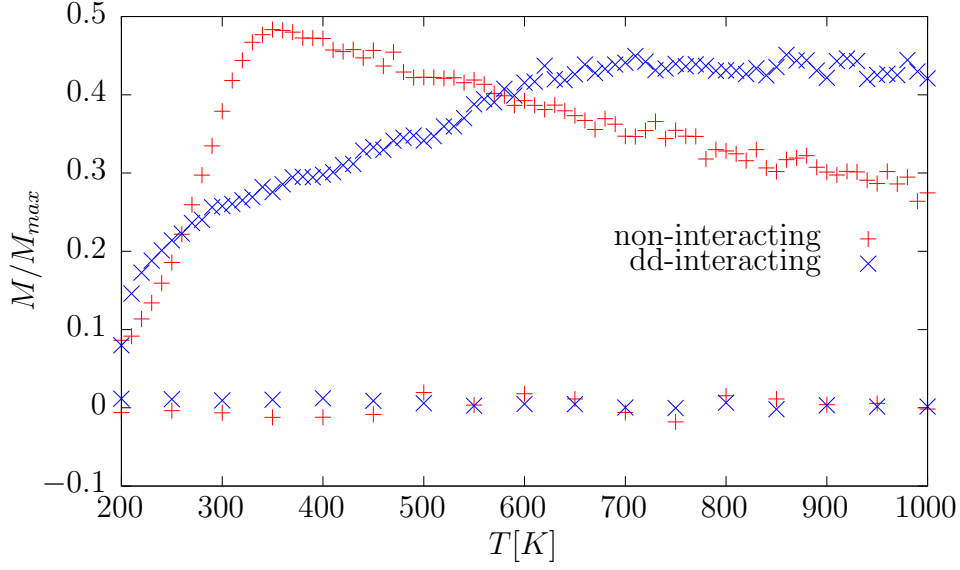


Figure 50: Simulated ZFC curve for maghemite nanoparticles at $\mu_0 H = 8mT$ with and without dipole-dipole interaction. The observed difference in T_B is highly exaggerated, as is the difference in shape. This is because the number of MCS is chosen too low and the blue curve does not represent a system that has found its thermal equilibrium at any point

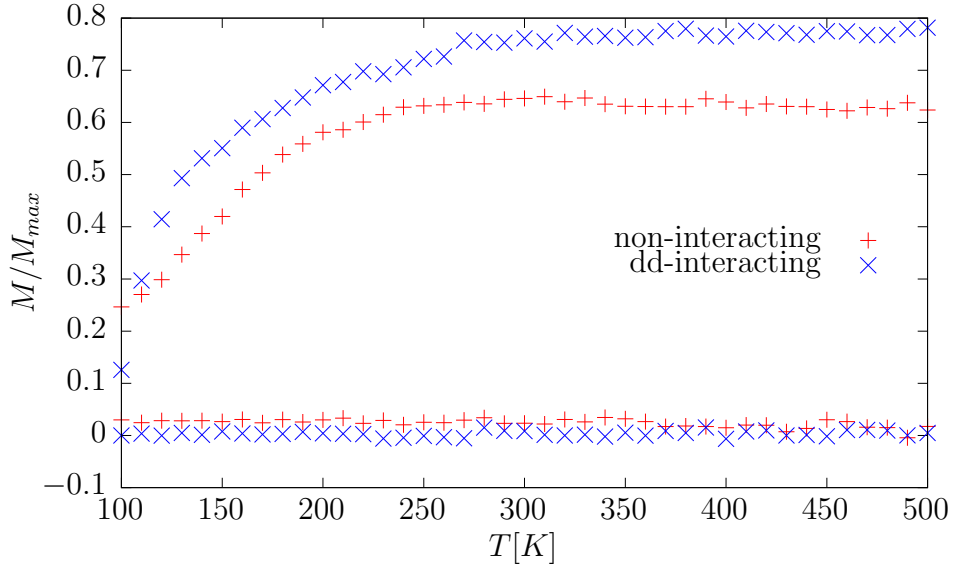


Figure 51: Simulated ZFC curve for maghemite nanoparticles at $\mu_0 H = 20mT$ with and without dipole-dipole interaction. In contrast to figure 50, the number of MCS per temperature step is increased by a factor of 10 with unaltered $d_m = 0.1$. The result is in far better accordance with experiments because the shape is better preserved and the difference in T_B is only at about 100K

6 Summary and Outlook

From the study of interacting particles, we have learned about the significance of simulation parameters like the test-vector length. The introduction of the Onsager Reaction Field method has been shown to be a valid approximation that can be used for magnetometric simulations like ZFC/FC curves in order to drastically reduce computational effort.

The presented research on purely dipolar systems in *fcc*-geometry has shown peculiar magnetic behavior that is very much distinct from magnetic phenomena associated with short-range exchange interactions. Our research was very much restricted only towards pure bulk behaviour. Certainly surface effects are extremely important in conjunction with dipole-dipole interaction and need to be addressed in a next step. Also it appears quite likely that the magnetocrystalline anisotropy has a large impact when studying systems with long-range magnetic order. Therefore, the limit of vanishing anisotropy is not a satisfactory basis for understanding realistic behaviour of assemblies of nanoparticles.

Because our current algorithm and implementation does allow for any choice in lattice constant, saturation magnetization, anisotropy distribution etc, future work should be done by studying larger regions of parameter space.

A Detailed Calculations

A.1 Magnetostatic Derivation of the Onsager Reaction Field

The ORF is a consequence of the magnetic Poisson equation that was derived, including suitable boundary conditions, in 5.1.6.2. Our 'magnetic sources' $\nabla \cdot \mathbf{M}$ are zero almost everywhere:

- Except for the magnetic dipole in the center, the inside of our cut-off sphere $\{\mathbf{r} \in \mathbb{R}^3 \mid 0 < r < R\}$ is modeled as vacuum, i.e. $\mathbf{M} = 0$.
- The outside $\{\mathbf{r} \in \mathbb{R}^3 \mid r > R\}$ is homogeneously magnetized, i.e. $\nabla \cdot \mathbf{M} = 0$.

Therefore, it is more elegant to use the general solution of the homogeneous Laplace equation which is easier to find, and account for everything else by finding special solutions at $r = 0$ and $r = R$ via the boundary conditions. Since the solution to any boundary value problem as stated is unique, any solution we find, will automatically be the correct solution.

A.1.1 Laplace Equation in Azimuthal Symmetry

We are interested in the solution to the homogeneous *Laplace equation* in spherical coordinates:

$$\Delta W(\mathbf{r}) = 0; \quad W(\mathbf{r}) = W(r, \theta, \varphi)$$

$$\frac{1}{r} \frac{\partial^2}{\partial r^2}(rW) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial W}{\partial \theta} \right) + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2 W}{\partial \varphi^2} = 0 \quad (\text{A.1})$$

We will immediately ignore the φ -part because we can always reduce our systems of interest to such with azimuthal symmetry. Furthermore, we make a separation ansatz:

$$W(r, \theta, \varphi) = W(r, \theta) = \frac{u(r)}{r} P(\theta)$$

Plugging this into (A.1) and multiplying by $r^2/(uP)$ yields

$$\underbrace{\frac{r^2}{u} \frac{d^2 u}{dr^2}}_{\text{independent from } \theta} + \underbrace{\frac{1}{P \sin \theta} \frac{d}{d\theta} \left(\sin \theta \frac{dP}{d\theta} \right)}_{\text{independent from } r} = 0 \quad (\text{A.2})$$

Therefore, both summands have to be simultaneously constant and opposite in sign. We therefore can define

$$\frac{r^2}{u} \frac{d^2 u}{dr^2} =: \lambda \in \mathbb{R} \quad \Rightarrow \quad \frac{d^2 u}{dr^2} - \frac{\lambda}{r^2} u = 0 \quad (\text{A.3})$$

and

$$x := \cos \theta \quad \Rightarrow \quad \frac{d}{dx} \left(\bullet \right) = -\frac{1}{\sin \theta} \frac{d}{d\theta} \left(\bullet \right) \quad (\text{A.4})$$

With these definitions we obtain from the second summand in (A.2) the ordinary differential equation

$$\begin{aligned} \frac{d}{dx} \left[(1-x^2) \frac{dP}{dx} \right] + \lambda P &= 0 \\ (1-x^2) \frac{d^2 P}{dx^2} - 2x \frac{dP}{dx} + \lambda P &= 0 \quad -1 \leq x \leq 1 \end{aligned} \quad (\text{A.5})$$

At this point, we must stress that the solutions need to cover the complete domain $x \in [-1, 1]$. We now define a function $w_n(x)$

$$w_n(x) := -(x^2 - 1)^n \quad (\text{A.6})$$

$$\Rightarrow (1-x^2)w'_n + 2nxw_n = 0$$

$$\text{Differentiate } n+1 \text{ times} \Rightarrow (1-x^2)w_n^{(2+n)} - 2xw_n^{(n+1)} + n(n+1)w_n^{(n)} = 0 \quad (\text{A.7})$$

This means that we have found functions w'_n that solve our 2nd order ordinary differential equation (A.5) including the necessary condition that it is defined $\forall x \in [-1, 1]$. These functions are therefore the only possible and unique solutions and we found that λ is not an arbitrary real number, but has to have the form

$$\lambda = n(n+1)$$

so that our differential equation is the *Legendre Differential Equation*

$$\frac{d}{dz} \left[(1-z^2) \frac{d}{dz} P_n(z) \right] + n(n+1)P_n(z) = 0$$

The so-called *Rodrigo form*

$$P_n := w'_n = \frac{d^n}{dx^n} (x^2 - 1)^n$$

that we recovered up to a normalization factor produces the *Legendre Polynomials* $P_n(x)$

$$\begin{aligned} P_0(x) &= 1 \\ P_1(x) &= x \\ P_2(x) &= \frac{1}{2}(3x^2 - 1) \\ P_3(x) &= \frac{1}{2}(5x^3 - 3x) \\ &\vdots \end{aligned}$$

Since we have also found what λ is, we can now investigate (A.3):

$$\frac{d^2 u}{dr^2} - \frac{n(n+1)}{r^2} u = 0;$$

The general solution to this for any given $n \in \mathbb{N}_0$ is

$$u(r) \equiv u_n(r) = a_n r^n + \frac{b_n}{r^{n+1}} \quad (\text{A.8})$$

Together, the general solution of the Laplace equation for problems with azimuthal symmetry is

$$W(\mathbf{r}) = W(r, \theta) = \sum_{l=0}^{\infty} \left(a_l r^l + \frac{b_l}{r^{l+1}} \right) P_l(\cos \theta) \quad (\text{A.9})$$

$$W(r) \text{ finite for } r = 0 \Rightarrow b_l = 0 \quad \forall l \geq 0$$

$$W(r) \rightarrow 0 \text{ for } r \rightarrow \infty \Rightarrow a_l = 0 \quad \forall l \geq 0$$

$$W(r) = A(r) \mathbf{a} \cdot \mathbf{r} \Rightarrow \nabla W(r) = \frac{d}{dr} A(r) \hat{\mathbf{r}} (\mathbf{a} \cdot \mathbf{r}) + \mathbf{a} A(r)$$

A.1.2 Solution to Our Boundary Value Problem

We restate the two distinct boundary value problems from section 5.1.6.2 with azimuthal symmetry that we want to solve with the above.

$$\Delta W^{\text{I/II}} = \nabla \cdot \mathbf{M}^{\text{I/II}}$$

$$W^{\text{I}}(r \rightarrow 0) \rightarrow \frac{\mathbf{m} \cdot \mathbf{r}}{4\pi r^3}$$

$$W^{\text{I}}(r \rightarrow \infty) \rightarrow 0$$

$$W^{\text{I}}(r = R) = \text{continuous}$$

$$\mathbf{H}_{in}^{\text{I},\perp} = \mu_r \mathbf{H}_{out}^{\text{I},\perp}$$

$$\mathbf{H}_{in}^{\text{I},\parallel} = \mathbf{H}_{out}^{\text{I},\parallel}$$

$$W^{\text{II}}(r \rightarrow 0) \rightarrow 0$$

$$W^{\text{II}}(r \rightarrow \infty) \rightarrow -\mathbf{H}_{ext} \cdot \mathbf{r}$$

$$W^{\text{II}}(r = R) = \text{continuous}$$

$$\mathbf{H}_{in}^{\text{II},\perp} = \mu_r \mathbf{H}_{out}^{\text{II},\perp}$$

$$\mathbf{H}_{in}^{\text{II},\parallel} = \mathbf{H}_{out}^{\text{II},\parallel}$$

Because the Legendre polynomials form an orthonormal set, one only needs to look at the powers of $\cos \theta$ that show up in the equations. This simplifies the problem to the following set of equations.

$$W_{\text{in}}^{\text{I}}(\mathbf{r}) = \alpha \frac{\cos \theta}{r^2} + \beta r \cos \theta$$

$$W_{\text{out}}^{\text{I}}(\mathbf{r}) = \gamma \frac{\cos \theta}{r^2} + \delta r \cos \theta$$

with $\alpha, \beta, \gamma, \delta \in \mathbb{R}$ to be determined from the boundary conditions.

B Programming

B.1 C++ implementation of Metropolis algorithm

Here we give the C++ implementation that has given all simulation results that were presented in this thesis, except for the dynamic susceptibility measurements. To achieve this versatility of one programme, the user must set logical parameters declaring what part of the programme will be relevant when executed. Also, the user can still declare things when running the executable. It is written in the C++11 standard.

The basic structure of this implementation is taken from an older C programme with permission by Oleg Petravic. Both the transition to the more modern C++ realization and the treatment of 2-particle interactions, especially within ORF were written by the author.

B.1.1 main.cpp

```

1  /*
2  MonteCarlo.cpp - Main file
3
4  M vs T / M vs B - simulation
5
6  !!!!
7  This programme is written in C++11 standard and utilizes openMP for easily implemented
   multithreaded computations
8  !!!!
9
10 The following is based on work leading to MnO Bulk, see its history below
11
12 Modification: 23/01/03, 24/01/03: main loop structure
13 Modification: 27/02/03: show+save Dip.- and Anis.-Energies -> MCS.one()
14 Modification: Feb/2009: rename to 5, with and without periodic boundary cond.
15 Modification: Nov/2012: MnO Bulk
16 START OF HEAVY MODIFICATION/REDUCTION since Nov/2016
17 Modification: Nov/2016: Transition to C++, intended to be executed on Linux -> e.g. substituting
   Visual C elements like '_s' functions.
18 Modification: Dec/2016: Heavy reduction leading to Heisenberg Paramagnetic simulation. Spin data
   as output for visualization in gnuplot or similar. Necessary dipole-dipole functions added
19 Modification: Jan/2017: sc lattice generator for easiest super-crystal structure. Optional cut-
   off implementation to speed up computation.
20 Modification: Feb/2017: include anisotropy energy. implement random distribution easy axes at
   every site
21 Modification: Sep/2017: Inclusion of perAux.h and introduction of ORF method for speedy
   simulations of interacting, periodic systems
22
23 */
24
25 #include <iostream>
26 #include <fstream>
27 #include <cmath>
28 #include <vector>
29 #include <array>
30 #include <string>
31 #include <algorithm>
32 #include <thread>
33
34 #include <stdlib.h>
35 #include <omp.h>
36
37 #include "parameter.h" // parameter file
38 #include "rnd250.c" // random number generation
39 #include "random_spd5.h" // rng addition
40 #include "input_ini.h" // necessary programming stuff like arrays, file save
41 #include "perAux.h" // contains functions for periodic boundary conditions and ORF method
42 #include "ORF0_str.h" // functions with physical meaning
43
44 using namespace std;

```

```

45
46 int main()
47 {
48     cout << "NN_estimate:_ " << NN << endl;
49     cout << "Include_d-d_interaction?_" ;
50     cin >> DIPOLAR;
51     int edition;
52     cout << "Edition:_ ";
53     cin >> edition;
54     cout << endl;
55     latt_const = latt_const_0;
56
57     int xx, StepNr, lx, lm, c;
58     xx = 1;
59     xxmax = 1;
60
61     reset_files(edition);
62
63     if (PERIODIC)
64     {
65         if (type == "sc")
66         {
67             sc_generator_new(edge_N);
68             max_N = pow(edge_N,3);
69         }
70         if (type == "bcc")
71         {
72             bcc_generator_new(edge_N);
73             max_N = 2*pow(edge_N,3);
74         }
75         if (type == "fcc")
76         {
77             fcc_generator_new(edge_N);
78             max_N = 4*pow(edge_N,3);
79         }
80
81         if (type == "hcp")
82         {
83             hcp_generator_new(edge_N);
84             max_N = 1*pow(edge_N,3);
85         }
86
87         makePeriodic();
88
89     }
90     else
91     {
92         if (type == "sc")
93             sc_generator_new(edge_N);
94         if (type == "bcc")
95             bcc_generator_new(edge_N);
96         if (type == "fcc")
97             fcc_generator_new(edge_N);
98         if (type == "hcp")
99             hcp_generator_new(edge_N);
100     if (SPHERICAL)
101     {
102         max_N = det_spheric();
103         new_pos();
104     }
105     else
106     {
107         max_N = max_N_temp;
108         cout << max_N << "_in_cube.\n";
109     }
110 }
111
112
113 InitMain(edition);
114
115 auto t1 = chrono::high_resolution_clock::now();
116
117 vector<double> comp_t;
118 comp_t.push_back(0.);
119 double Et = 0.;
120
121 // ***** Scans *****
122
123 for (int cfg = 0; cfg < CFG; cfg++) // config loop
124 {
125
126     InitArray();

```

```

127     anisosave(edition);
128     aniso_test();
129
130     cout << "start_T_sweep\n";
131     // ***** T-Scan *****
132     T[xx] = Tpt[0];
133     Bx[xx] = Bxfix[0];
134     //BBx[xx] = Cz * Bx[xx] // OFF In para0 not necessary
135
136     for (c = 0; c < MR; c++) // sweep counter loop
137     {
138
139
140         // one single temperature step at the beginning
141         if (Tsteps[c] > 0)
142         {
143             T[xx] = Tpt[c];
144             Bx[xx] = Bxfix[c];
145             array<double, 2> arr = rel_perm(Bx[xx]);
146             double mu_r = arr[0];
147             double M = arr[1];
148             for (lx = 0; lx < TRlxLoops[c]; lx++) // Relaxation loop
149             {
150                 MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
151             }
152
153             for (lm = 0; lm < TAvGLoops[c]; lm++) // Averaging loop
154             {
155                 MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
156                 Mx[xx] += measure();
157             }
158
159             Mx[xx] /= ((double) TAvGLoops[c]);
160
161
162             auto t2 = chrono::high_resolution_clock::now();
163
164             chrono::duration<double, milli> fp_ms = t2-t1;
165             comp_t.push_back(fp_ms.count());
166
167             spinsave(edition); // NOT advised here! GB files!
168             filesave(edition, xx);
169             double Et = E_filesave(edition, xx);
170             detailed_measure(edition, xx);
171
172             cout << "cfg:_ " << cfg + 1 << ",_T=_ " << T[xx] << ",_B=_ " <<
173                 Bx[xx] << ",_Mx=_ " << Mx[xx] << ",_Etot=_ " << Et << "_in_"
174                 ;
175             cout << (comp_t[xx]-comp_t[xx-1])/1000 << "_[s]\n";
176
177             cout << "rel_Permeability:_ " << mu_r;
178             if (mu_r < 1)
179             {
180                 cout << "_>_physical?!";
181                 //abort();
182             }
183             cout << endl;
184
185
186             xx++;
187             xxmax++;
188
189             T[xx] = T[xx-1] + dT[c];
190
191         }
192
193
194         for (StepNr = 0; StepNr < Tsteps[c]; StepNr++)
195         {
196             Bx[xx] = Bxfix[c];
197             array<double, 2> arr = rel_perm(Bx[xx]);
198             double mu_r = arr[0];
199             double M = arr[1];
200             for (lx = 0; lx < TRlxLoops[c]; lx++) // Relaxation loop
201             {
202                 MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
203             }
204
205
206             for (lm = 0; lm < TAvGLoops[c]; lm++) // Averaging loop

```

```

207         {
208             MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
209             Mx[xx] += measure();
210         }
211
212     Mx[xx] /= ((double) TAvLoops[c]);
213
214
215     auto t2 = chrono::high_resolution_clock::now();
216
217     chrono::duration<double, milli> fp.ms = t2-t1;
218     comp.t.push_back(fp.ms.count());
219
220     spinsave(edition); // NOT ADVISED: GB files!
221     filesave(edition, xx);
222     Et = E_filesave(edition, xx);
223     detailed_measure(edition, xx);
224
225     cout << "cfg:_ " << cfg + 1 << ",_T=_ " << T[xx] << ",_B=_ " <<
        Bx[xx] << ",_Mx=_ " << Mx[xx] << ",_Etot=_ " << Et << "_in_"
        ;
226     cout << (comp.t[xx]-comp.t[xx-1])/1000 << "_[s]\n";
227
228
229     cout << "rel_Permeability:_ " << mu_r;
230     if (mu_r < 1)
231     {
232         cout << "_->_physical?!";
233         //abort();
234     }
235     cout << endl;
236
237     //cout << "spin update\n";
238
239     xx++;
240     xxmax++;
241
242     T[xx] = T[xx-1] + dT[c];
243
244 }
245
246
247 }
248
249 // cout << "start B sweep\n";
250 // ***** B-Scan ***** // Won't be activated here as Tsteps[] =
    0;
251
252 Bx[xx] = Bxpt[0];
253 T[xx] = Tfix[0];
254
255 for (c = 0; c < MR; c++)
256 {
257     // one single field step at the beginning
258     // int status = 0; // variable 'status' and function 'countdown'
        are strictly cosmetic: ~ computation status bar
259     if (Bsteps[c] > 0)
260     {
261         Bx[xx] = Bxpt[c];
262         T[xx] = Tfix[c];
263         array<double, 2> arr = rel.perm(Bx[xx]);
264         double mu_r = arr[0];
265         double M = arr[1];
266         //cout << "Single step, relaxation: ";
267
268         for (lx = 0; lx < BRlxLoops[c]; lx++) // Relaxation loop
269         {
270             MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
271             //status = countdown(BRlxLoops[c],
                lx, status);
272         }
273
274         for (lm = 0; lm < BAvLoops[c]; lm++) // Averaging loop
275         {
276             MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
277             Mx[xx] += measure();
278         }
279
280         Mx[xx] /= ((double) TAvLoops[c]);
281
282         auto t2 = chrono::high_resolution_clock::now();

```

```

283         chrono::duration<double, milli> fp.ms = t2-t1;
284         comp.t.push_back(fp.ms.count());
285
286         spinsave(edition);          // NOT ADVISED: GB files!
287         filesave(edition, xx);
288         Et = E.filesave(edition, xx);
289         detailed_measure(edition, xx);
290
291         cout << "cfg:_ " << cfg + 1 << ",_T=_ " << T[xx] << ",_B=_ " <<
                Bx[xx] << ",_Mx=_ " << Mx[xx] << ",_Etot=_ " << Et << "_in_"
                ;
292         cout << (comp.t[xx]-comp.t[xx-1])/1000 << "_[s]\n";
293
294
295
296         ++xx;
297         ++xxmax;
298
299         Bx[xx] = Bx[xx-1] + dBx[c];
300     }
301
302     //status = 0;
303     for (StepNr = 0; StepNr < Bsteps[c]; StepNr++)
304     {
305         T[xx] = Tfix[c];
306         array<double, 2> arr = rel_perm(Bx[xx]);
307         double mu_r = arr[0];
308         double M = arr[1];
309         //cout << "Relaxation progress: ";
310         for (lx = 0; lx < BRlxLoops[c]; lx++) // Relaxation loop
311         {
312             MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
313             //status = countdown(BRlxLoops[c], lx, status);
314
315         }
316
317         for (lm = 0; lm < BAvgLoops[c]; lm++) // Averaging loop
318         {
319             //cout << "Schleife\n";
320
321             MCS_one(xx, Bx[xx], KB*T[xx], mu_r, M);
322             Mx[xx] += measure();
323             //status = countdown(BAvgLoops[c], lm, status);
324         }
325
326         Mx[xx] /= ((double) TAvgLoops[c]);
327         //cout << "Averaging done\n";
328
329         auto t2 = chrono::high_resolution_clock::now();
330
331         chrono::duration<double, milli> fp.ms = t2-t1;
332         comp.t.push_back(fp.ms.count());
333
334         spinsave(edition);          // NOT ADVISED: GB files!
335         filesave(edition, xx);
336         Et = E.filesave(edition, xx);
337         detailed_measure(edition, xx);
338
339         cout << "cfg:_ " << cfg + 1 << ",_T=_ " << T[xx] << ",_B=_ " <<
                Bx[xx] << ",_Mx=_ " << Mx[xx] << ",_Etot=_ " << Et << "_in_"
                ;
340         cout << (comp.t[xx]-comp.t[xx-1])/1000 << "_[s]\n";
341
342
343         ++xx;
344         ++xxmax;
345
346         Bx[xx] = Bx[xx-1]+dBx[c];
347     }
348
349     // output.dat only updated after 1 sweep, NOT after every step as spin.
    dat !
350
351
352
353
354
355     // OFF spintable();
356
357 } // \configuration loop
358
359

```

```

360     }
361     return 0;
362 }
363
364
365 //end.

```

B.1.2 parameter.h

```

1  // parameter.h
2  // configuration file
3  // for MC simulation
4
5  using namespace std;
6
7
8  extern const string data_file = "output";
9  extern const string table_file = "spin";
10 extern const string aniso_file = "aniso";
11 extern const string Edata_file = "Edata";
12 extern const string Mdata_file = "Mdata";
13 //extern const string blockT_file = "blockT.dat";
14
15
16 // Natural constants. Only to be changed for convenience!!
17 const double pi = M_PI; // pi
18 const double KB = 1.38064852e-23; // Boltzmann constant
19 const double mu0 = pi*4e-7; // vacuum permeability
20 const double eV = 1.60217662e-19; // electron charge for easy conversion Joule <-> electron Volt
21 const double hbar = 1.0545718e-34; // h bar
22
23 // Parameters that distinguish the sample/experiment and its dynamics
24 const double dm = 1.; // test-rotation vector length
25 const double Mag = 0.38e6*pi/6*pow(20e-9,3); // Magnetic moment (saturation mag. * Volume)
26 //const double KV = 0.;
27 const double KV = 1.34e4*pi/6*pow(20e-9,3); // anisotropy constant * Volume //
28 const double latt_const_0 = 1.*20e-9*1.1*sqrt(2.); // lattice constant of outer sc lattice
29 double latt_const; // lattice updated via edition parameter!!
30 const double def_mur = 15.;
31
32
33
34 // Parameters that define size of system and thus largely influence computation time!
35 //const bool DIPOLAR = true;
36 bool DIPOLAR;
37 const bool PERIODIC = true; // Calculate periodic system with ORF approx. If true, SPHERICAL is
    inactive
38 const bool PRINT_ENERGIES = false; // if true, all total energies are regularly calculated and
    saved
39 const bool PRINT_SPINS = false; // if true, spin-tables are regularly printed (GB files!!)
40 const bool DETAILED_MEASURE = true; // if true, other magnetizations than mx and their abs are
    recorded
41 const bool ONSDAT = false; // if true, all (!!!) Onsager energies will be recorded -> GB files!!
42 extern const string type = "fcc";
43 const bool SPHERICAL = false; // if true, original cube is cut off to a sphere
44
45 const int edge_N = 7; // # spins per edge
46 //const int max_N_temp = 2046 // # spins being considered
47 //const int max_N_temp = pow(edge_N,3); // general sc
48 int max_N;
49 //const int max_N_temp = pow(edge_N,3) + pow(edge_N-1,3); // general bcc
50 const int max_N_temp = 4*pow(edge_N,3); // general fcc
51 //const int max_N_temp = pow(edge_N,3) + 3*edge_N*pow(edge_N-1,2);
52 const int loopNrAvg = 10; // # MC Avgloops per step
53 const int loopNrRlx = 500; // # MC Rlxloops per step // !! in anisoBlock: loopNr2 = loopNr
    always !!
54 const double CUTOFF = 2; // maximum distance up to which dipole energy is calculated
55 double BEST_CUTOFF; // largest radius containing same neighbours as above value
56
57
58 // Parameters with mostly computational rather than physical meaning
59 const int numThreads = 4; // number of possible parallel threads
60 const int numThreads2 = 2; // alternative number of parallel threads
61 //const int Ntrunc = max_N/num_threads;
62 const int SEED = 11102017; //date as seed for rnd250()
63 const int MR = 3; // number of possible sweeps
64 const int MAXDATA = 100000; // max. number of data points
65 const int CFG = 1; // number of configurations
66
67
68 // Temperature T in K(elvin)

```

```

69 // and
70 // magnetic field B in T(esla)
71
72 // Points for T - sweeps
73 const double Tpt[MR+1] = {1000, 25, 1000, 1000};
74 const double dT[MR] = {-25, 25, -100};
75 const double Bxfix[MR] = {0.0, 0.015, 0.015};
76 const int TRlxLoops[MR] = {loopNrRlx, loopNrRlx, loopNrRlx};
77 const int TAvlLoops[MR] = {loopNrAvg, loopNrAvg, loopNrAvg};
78
79 // Points for B- sweeps
80 const double Bxpt[MR+1] = {0.15, 0.15, 0.15, 0.15};
81 const double dBx[MR] = {0.005, -0.005, 0.005};
82 const double Tfix[MR] = {10, 10, 10};
83 const int BRlxLoops[MR] = {loopNrRlx, loopNrRlx, loopNrRlx};
84 const int BAvlLoops[MR] = {loopNrAvg, loopNrAvg, loopNrAvg};

```

B.1.3 input-ini.h

```

1 // input-ini.h
2
3 // based on ini_spd5tbl.h
4 // computational necessities without physical meaning are defined here.
5 //
6
7
8 using namespace std;
9
10
11 // Spin positions
12 double POS[max_N_temp][3]; // Generates the sc lattice of spin positions.
13 bool intern[max_N_temp]; // flags if sites are within spherical cutoff
14 double N_POS[max_N_temp][3];
15
16
17 // Spin tables
18 double s[max_N_temp][3];
19 double sx[max_N_temp], sy[max_N_temp], sz[max_N_temp];
20
21 // Anisotropy easy axes
22 double k[max_N_temp][3];
23 double kx[max_N_temp], ky[max_N_temp], kz[max_N_temp];
24
25
26 void ini_config(); // initialize configuration
27 void InitArray(); // array initialization function -> CFG loop
28 void dist_matrix(); // calculate distance matrix for spin interaction
29 void r_vectors(); // calculate normalized distance vectors
30 void local_matrix(); // determine neighbourhood positions for dipole interaction
31 void line_generator(); // generates line of positions
32 void sc_generator(); // sc generator
33 void bcc_generator(); // bcc generator
34 void fcc_generator(); // fcc generator
35 void sc_only(); // POS only with sc generated
36 double best_cutoff();
37
38 int Tsteps[MR], Bsteps[MR];
39 int xxmax;
40 double T[MAX_DATA], Bx[MAX_DATA];
41 double Mx[MAX_DATA];
42 double TBsteps[CFG][5];
43
44
45
46
47
48 void InitMain(int ed)
49 {
50     int maxdat, i, xx;
51     int SEED_cluster;
52     if (ed < 1000)
53         SEED_cluster = SEED ;
54     else
55         SEED_cluster = SEED + ed;
56     seed250(SEED_cluster);
57     srand(SEED_cluster);
58
59     maxdat = 0;
60     for (i = 0; i < MR; i++)
61     {
62         //Tsteps[i] = abs( (int) ((Tpt[i]-Tpt[i+1])/dT[i]));

```

```

63         // Pure B Sweep !!!
64         Tsteps[i] = abs( (int) ((Tpt[i]-Tpt[i+1])/dT[i]));
65         Bsteps[i] = abs( (int) ((Bxpt[i]-Bxpt[i+1])/dBx[i]));
66         //Bsteps[i] = 0; // Only T sweeps here !!
67         maxdat += Tsteps[i]+Bsteps[i];
68     }
69
70     cout << (maxdat+1)*CFG << "_Data_points\n";
71
72     if ((maxdat*CFG+1) > MAX.DATA)
73     {
74         cout << "too_many_data_points!!\n";
75         abort();
76     }
77
78     for (xx = 0; xx < MAX.DATA; xx++)
79     {
80         Mx[xx] = 0.0;
81         T[xx] = 0.0;
82         Bx[xx] = 0.0;
83     }
84     for (int i = 0; i < CFG; i++)
85     {
86         for (int j = 0; j <=5; j++)
87         {
88             TBsteps[i][j] = 0;
89         }
90     }
91
92     cout << "data_variables_initialized!\n";
93 }
94
95 void InitArray()
96 {
97
98     cout << "initialize_arrays...\n";
99     ini_config();
100     cout << "arrays_initialized!\n";
101
102     // If no dipole-dipole interaction is considered, below functions are NOT necessary
103
104     cout << "calculate_distances...\n";
105     dist_matrix();
106     cout << "distances_calculated!\n";
107
108     if (PERIODIC)
109         BEST.CUTOFF = best_cutoff();
110
111     if (PRINT.ENERGIES or not PERIODIC)
112     {
113
114         cout << "calculate_distance_vectors...\n";
115         r_vectors();
116         cout << "distance_vectors_calculated!\n";
117
118     }
119
120
121
122     /*
123     cout << "calculate dipole neighbours\n";
124     local_matrix();
125     cout << "dipole neighbours determined\n";
126     */
127 }
128
129 void ini_config()
130 {
131
132     for (int i = 0; i < max.N; i++)
133     {
134         // Both spin and easy axis are stored as unit vectors because we need only the
135         // angle between them via scalar product
136
137         // Set initial spin vector directions
138         Marsaglia(s[i]);
139
140         sx[i] = s[i][0];
141         sy[i] = s[i][1];
142         sz[i] = s[i][2];
143
144         // Set initial easy axes directions

```

```

144         Marsaglia(k[i]);
145
146         kx[i] = k[i][0];
147         ky[i] = k[i][1];
148         kz[i] = k[i][2];
149
150
151
152     }
153     cout << "Initial_spin_directions_set.\n";
154     cout << "Easy_axes_are_set.\n";
155 }
156
157
158 // Two optional functions that are only useful when comparing magnetization to theoretical
    paramagnetic case!
159
160 double Langevin(double Ezee, double kbT)
161 {
162     return (1/tanh(Ezee/kbT)-kbT/Ezee);
163 }
164
165 // Reset all output files
166 void reset_files(int ed)
167 {
168     string data_file_cluster = data_file + to_string(ed) + ".dat";
169     string table_file_cluster = table_file + to_string(ed) + ".dat";
170     string aniso_file_cluster = aniso_file + to_string(ed) + ".dat";
171     string Edata_file_cluster = Edata_file + to_string(ed) + ".dat";
172     string Mdata_file_cluster = Mdata_file + to_string(ed) + ".dat";
173
174     ofstream fout;
175     fout.open(data_file_cluster, ios::trunc);
176     fout.close();
177
178     ofstream spinout;
179     spinout.open(table_file_cluster, ios::trunc);
180     spinout.close();
181
182     ofstream anisout;
183     anisout.open(aniso_file_cluster, ios::trunc);
184     anisout.close();
185
186     ofstream Efout;
187     Efout.open(Edata_file_cluster, ios::trunc);
188     Efout.close();
189
190     ofstream Mfout;
191     Mfout.open(Mdata_file_cluster, ios::trunc);
192     Mfout.close();
193
194     if (ONSDAT)
195     {
196         ofstream ons;
197         ons.open("ons.dat", ios::trunc);
198         ons.close();
199     }
200
201     /*
202     ofstream blockout;
203     blockout.open(blockT_file, ios::trunc);
204     blockout.close();
205     */
206 }
207
208
209 // Save data_file which contains magnetization at applied field, temperature
210 void filesave(int ed, int xx)
211 {
212     string data_file_cluster = data_file + to_string(ed) + ".dat";
213     ofstream fout;
214     fout.open(data_file_cluster, ios::app);
215     //fout << "cgf T B M\n";
216
217     fout << xx << "\t" << T[xx] << "\t" << Bx[xx] << "\t" << Mx[xx] << endl;
218
219
220     fout.close();
221 }
222
223
224 // Save spin positions. Every block of max_N rows corresponds to one row in data_file.

```

```

225 void spinsave(int ed)
226 {
227     if (PRINT.SPINS)
228     {
229
230         string table_file_cluster = table_file + to_string(ed) + ".dat";
231
232         ofstream spinout;
233         spinout.open(table_file_cluster, ios::app);
234         //fout << "cgf T B M\n";
235
236         double X,Y,Z,VX,VY,VZ;
237
238
239
240         for (int n = 0; n < max.N; n++)
241         {
242
243
244             VX = sx[n];
245             VY = sy[n];
246             VZ = sz[n];
247
248             X = POS[n][0];
249             Y = POS[n][1];
250             Z = POS[n][2];
251
252             /*
253
254             // Operations on sx, POS in order to make the plots in gnuplot
255                 easier
256             VX = 0.5*sx[n];
257             VY = 0.5*sy[n];
258             VZ = 0.5*sz[n];
259
260             X = POS[n][0]/latt_const - 0.5*VX;
261             Y = POS[n][1]/latt_const - 0.5*VY;
262             Z = POS[n][2]/latt_const - 0.5*VZ;
263             */
264             spinout << X << "\t" << Y << "\t" << Z << "\t" << VX << "\t" <<
265                 VY << "\t" << VZ << endl;
266
267             //spinout << POS[n][0] << "\t" << POS[n][1] << "\t" << POS[n][2]
268                 << "\t" << sx[n] << "\t" << sy[n] << "\t" << sz[n] << endl;
269
270         }
271         //fout << endl;
272
273         spinout.close();
274     }
275 }
276
277 // Save easy axes at every site analogously to spinsave.
278 void anisosave(int ed)
279 {
280
281     string aniso_file_cluster = aniso_file + to_string(ed) + ".dat";
282     ofstream anisout;
283     anisout.open(aniso_file_cluster);
284
285     double X,Y,Z,VX,VY,VZ;
286
287
288
289     for (int n = 0; n < max.N; n++)
290     {
291
292
293         VX = kx[n];
294         VY = ky[n];
295         VZ = kz[n];
296
297         X = POS[n][0];
298         Y = POS[n][1];
299         Z = POS[n][2];
300
301         /*
302
303         VX = 0.5*kx[n];
304         VY = 0.5*ky[n];
305         VZ = 0.5*kz[n];
306
307         X = POS[n][0]/latt_const - 0.5*VX;
308         Y = POS[n][1]/latt_const - 0.5*VY;

```

```

304         Z = POS[n][2]/latt_const-0.5*VZ;
305         */
306
307         anisout << X << "\t" << Y << "\t" << Z << "\t" << VX << "\t" << VY << "\t"
308             << VZ << endl;
309
310         //spinout << POS[n][0] << "\t" << POS[n][1] << "\t" << POS[n][2] << "\t"
311             << sx[n] << "\t" << sy[n] << "\t" << sz[n] << endl;
312     }
313     //fout << endl;
314     cout << "Wrote_easy_axes_table.\n";
315     anisout.close();
316 }
317
318
319 //end.

```

B.1.4 str.h

```

1  // str.h
2  // functions and definitions that translate the physical structure of the simulated system are
   done here
3
4  using namespace std;
5
6
7
8  double dist[max_N_temp][max_N_temp];
9  double rVectors[max_N_temp][max_N_temp][3];
10
11 // function that calculates array dist[max_N][max_N]
12
13 void line_generator(int edge, double start[3], int direction, int start_index)
14 {
15     int n = start_index;
16     double a = 1.;
17     int dir[3] = {0,0,0};
18     dir[direction] = 1;
19
20     for (int i = 0; i < edge; i++)
21     {
22         POS[n][0] = a*i*dir[0]+start[0];
23         POS[n][1] = a*i*dir[1]+start[1];
24         POS[n][2] = a*i*dir[2]+start[2];
25         n++;
26     }
27 }
28
29 void sc_generator(int edge, double start[3], int start_index)
30 {
31     int n = start_index;
32     double a = 1.;
33
34     for (int i = 0; i < edge; i++)
35     {
36         for (int j = 0; j < edge; j++)
37         {
38             for (int k = 0; k < edge; k++)
39             {
40                 POS[n][0] = a*i+start[0];
41                 POS[n][1] = a*j+start[1];
42                 POS[n][2] = a*k+start[2];
43                 n++;
44             }
45         }
46     }
47 }
48
49 void sc_only(int outer_edge)
50 {
51     double sc_start[3] = {0,0,0};
52     sc_generator(outer_edge, sc_start, 0);
53     if (max_N_temp != pow(edge_N,3))
54     {
55         cout << "Error_in_sc_generation ,_check_site_numbers!" << endl;
56         abort();
57     }
58 }
59

```

```

60 void bcc_generator(int outer_edge)
61 {
62     double a = 1.;
63     double outer_start[3] = {0,0,0};
64     double inner_start[3] = {0.5*a,0.5*a,0.5*a};
65     if (outer_edge < 2 or max_N_temp != pow(edge_N,3)+pow(edge_N-1,3))
66     {
67         cout << "error_in_bcc_generation ,_check_site_numbers!" << endl;
68         abort();
69     }
70     sc_generator(outer_edge, outer_start, 0);
71     int outer_index = pow(outer_edge,3);
72     sc_generator(outer_edge-1, inner_start, outer_index);
73 }
74 }
75
76 void fcc_generator(int outer_edge)
77 {
78     double a = 1.;
79     double outer_start[3] = {0,0,0};
80
81     if (outer_edge < 2 or max_N_temp != pow(edge_N,3)+3*edge_N*pow(edge_N-1,2))
82     {
83         cout << "error_in_fcc_generation ,_check_site_numbers!" << endl;
84         abort();
85     }
86     sc_generator(outer_edge, outer_start, 0);
87     int current_index = pow(outer_edge,3);
88
89
90     // generate lines in x-direction
91     double current_start[3] = {0,0,0};
92
93     for (int i = 0; i < outer_edge-1; i++)
94     {
95         for (int j = 0; j < outer_edge-1; j++)
96         {
97             current_start[1] = a*(0.5+i);
98             current_start[2] = a*(0.5+j);
99             line_generator(outer_edge, current_start, 0, current_index);
100             current_index += outer_edge;
101         }
102     }
103     current_start[1] = 0;
104     current_start[2] = 0;
105
106     // generate lines in y-direction
107
108
109     for (int i = 0; i < outer_edge-1; i++)
110     {
111         for (int j = 0; j < outer_edge-1; j++)
112         {
113             current_start[0] = a*(0.5+i);
114             current_start[2] = a*(0.5+j);
115             line_generator(outer_edge, current_start, 1, current_index);
116             current_index += outer_edge;
117         }
118     }
119
120     current_start[0] = 0;
121     current_start[2] = 0;
122
123     // generate lines in z-direction
124
125
126     for (int i = 0; i < outer_edge-1; i++)
127     {
128         for (int j = 0; j < outer_edge-1; j++)
129         {
130             current_start[0] = a*(0.5+i);
131             current_start[1] = a*(0.5+j);
132             line_generator(outer_edge, current_start, 2, current_index);
133             current_index += outer_edge;
134         }
135     }
136
137     current_start[0] = 0;
138     current_start[1] = 0;
139
140
141

```

```

142 }
143
144 array<double, 4> calc_cent_rad()
145 {
146     array<double, 4> CENTER = {0,0,0,0};
147     for (int i = 0; i < max_N_temp; i++)
148     {
149         CENTER[0] += POS[i][0];
150         CENTER[1] += POS[i][1];
151         CENTER[2] += POS[i][2];
152     }
153
154     CENTER[0] /= max_N_temp;
155     CENTER[1] /= max_N_temp;
156     CENTER[2] /= max_N_temp;
157
158     CENTER[3] = 0.5 * (edge_N-1);
159     return CENTER;
160 }
161
162 bool in_sphere(double x, double y, double z, array<double,4> center)
163 {
164     double x2 = center[0];
165     double y2 = center[1];
166     double z2 = center[2];
167     double R = center[3];
168
169     double r = pow(x-x2,2) + pow(y-y2,2) + pow(z-z2,2);
170
171     return (r <= R*R);
172 }
173
174
175
176 int det_spheric()
177 {
178     int MAX = 0;
179
180     array<double,4> CENTER = calc_cent_rad();
181
182     //cout << "Center: " << CENTER[0] << " " << CENTER[1] << " " << CENTER[2] << endl;
183     //cout << "Radius: " << CENTER[3] << endl;
184
185     fill_n(intern, max_N_temp, false);
186
187
188     double x, y, z;
189
190     for (int i = 0; i < max_N_temp; i++)
191     {
192
193         x = POS[i][0];
194         y = POS[i][1];
195         z = POS[i][2];
196
197         //cout << x << " " << y << " " << z << endl;
198
199         if (in_sphere(x, y, z, CENTER))
200         {
201             MAX++;
202             intern[i] = true;
203         }
204     }
205
206 }
207
208 cout << MAX << "_of_" << max_N_temp << "_in_sphere.\n";
209
210 return MAX;
211 }
212
213
214 void new_pos()
215 {
216     int n = 0;
217
218     for (int i = 0; i < max_N_temp; i++)
219     {
220         if (intern[i])
221         {
222             N.POS[n][0] = POS[i][0];
223             N.POS[n][1] = POS[i][1];

```

```

224         N.POS[n][2] = POS[i][2];
225
226         n++;
227     }
228
229 }
230
231 if (n != max_N)
232 {
233     cout << "Error_in_spherical_cutoff!\n";
234     abort();
235 }
236
237 // reset POS with arbitrary value (-42)
238 fill(POS[0], POS[0] + max_N_temp * 3, -42.);
239
240 // for relevant indices, fill POS with N.POS
241 for (int i = 0; i < max_N; i++)
242 {
243     POS[i][0] = N.POS[i][0];
244     POS[i][1] = N.POS[i][1];
245     POS[i][2] = N.POS[i][2];
246 }
247
248
249
250
251 }
252 /*
253     const int MAX = max_N;
254
255     double N_POS[MAX][3];
256
257 */
258
259 void dist_matrix()
260 {
261     int i = 0;
262
263     while(i < max_N)
264     {
265         int j = i;
266         while (j < max_N)
267         {
268             dist[i][j] = latt_const * sqrt(pow(POS[i][0] - POS[j][0], 2) + pow(POS[i]
269                 ][1] - POS[j][1], 2) + pow(POS[i][2] - POS[j][2], 2));
270             j++;
271         }
272         i++;
273     }
274     int j = 0;
275     // counter loop for case i > j
276     while(j < max_N)
277     {
278         int i = j;
279         while (i < max_N)
280         {
281             dist[i][j] = dist[j][i];
282             i++;
283         }
284         j++;
285     }
286     //cout << "Test dist " << dist[2][53] << " " << dist[53][2] << endl;
287 }
288
289
290 // calculation of the normalized distance vectors between sites -> rVectors[max_N][max_N][3]
291 void r_vectors()
292 {
293     for (int i = 0; i < max_N; i++)
294     {
295         for (int j = i+1; j < max_N; j++)
296         {
297             //double d = dist[i][j];
298             double vec[3] = {POS[i][0] - POS[j][0], POS[i][1] - POS[j][1], POS[i][2] - POS[j]
299                 ][2]};
300             double mod = sqrt(pow(vec[0], 2) + pow(vec[1], 2) + pow(vec[2], 2));
301             vec[0] /= mod;
302             vec[1] /= mod;
303             vec[2] /= mod;

```

```

304
305         rVectors[i][j][0] = vec[0];
306         rVectors[i][j][1] = vec[1];
307         rVectors[i][j][2] = vec[2];
308         //cout << vec[2] << endl;
309
310         //Ed += 1./pow(d,3) * (3* (sx[j]*vec[0]+sy[j]*vec[1]+sz[j]*vec[2])*(sx[i]
           ]*vec[0]+sy[i]*vec[1]+sz[i]*vec[2]) - (sx[j]*sx[i]+sy[j]*sy[i]+sz[j]
           ]*sz[i]));
311     }
312
313 }
314
315
316 int j = 0;
317 // counter loop for case i>j
318 while(j < max.N)
319 {
320     int i = j;
321     while (i<max.N)
322     {
323         rVectors[i][j][0] = rVectors[j][i][0];
324         rVectors[i][j][1] = rVectors[j][i][1];
325         rVectors[i][j][2] = rVectors[j][i][2];
326         i++;
327     }
328     j++;
329 }
330
331 //cout << "Test r-Vec: " << rVectors[2][53][0] << " " << rVectors[2][53][1] << " " <<
           rVectors[2][53][2] << " " << rVectors[53][2][0] << " " << rVectors[53][2][1] << " "
           << rVectors[53][2][2] << endl;
332 }
333
334
335 // get the neighbours that are within the cutoff range and store their indices.
336 // Note that only i=0 gets all neighbours explicitly, neighbours for i>0 are partly contained
           for smaller i !!
337 /*
338 void local_matrix()
339 {
340     int min = max.N;           // completely optional:
341     int max = 0;               // show the minimal/maximal number of neighbours that will be
           considered for E-dipole
342
343     // loop over all sites i
344     for (int i = 0; i < max.N; i++)
345     {
346         int loc = 1;           // counts # neighbours for site i
347
348         loc_POS[i][0] = 0;      // 0th element contains number of neighbours, used in
           loc_E-dipole()
349
350         // because of symmetry of E-dipole, only upper triangle matrix needs to be filled
351         for (int j = i+1; j < max.N; j++) // only viable if loc_E-dipole() works with
           corresponding structure!
352         {
353             // loc_POS only filled with neighbouring sites within a distance of '
           cutoff' around site i
354             if (dist[i][j] <= cutoff)
355             {
356                 loc_POS[i][loc] = j; // So, loc_POS[a][b] = c reads: the
           position vector of the bth neighbour of site a is stored as
           cth element of the ORIGINAL POS array.
357                 loc++;
358                 loc_POS[i][0]++;
359             }
360             // Because of this construction, no dipole-dipole pair will be either
           counted double or forgotten!
361         }
362         if (loc > max)
363         {
364             max = loc;
365         }
366         if (loc < min)
367         {
368             min = loc;
369         }
370     }
371 }
372 }
373

```

```

374         cout << "Up to " << max-1 << " neighbours per site considered instead of original " <<
           max.N-1 << endl;
375     }
376     */
377
378     double dipole(int i, int j)
379     {
380         double d = dist[i][j];
381
382         double vec[3];
383         vec[0] = rVectors[i][j][0];
384         vec[1] = rVectors[i][j][1];
385         vec[2] = rVectors[i][j][2];
386
387         if (i!=j)
388             return -mu0*Mag*Mag/(4*PI * pow(d,3)) * (3* (sx[j]*vec[0]+sy[j]*vec[1]+sz[j]*vec
               [2])*(sx[i]*vec[0]+sy[i]*vec[1]+sz[i]*vec[2]) - (sx[j]*sx[i]+sy[j]*sy[i]+sz[
               j]*sz[i]));
389
390         else
391             return 0;
392     }
393
394     // this function calculates the sum of all dipole-dipole energies w/o cutoff!
395     double E_dipole()
396     {
397         double Ed = 0;
398         //int status;
399
400         for (int i = 0; i<max.N; i++)
401         {
402             for (int j = i+1; j<max.N; j++) // upper triangle matrix: main diagonal entries
403                 // must be 0, rest isn't determined because dipole pairs mustn't be counted
404                 // twice
405             {
406                 double d = dist[i][j];
407
408                 double vec[3];
409                 vec[0] = rVectors[i][j][0];
410                 vec[1] = rVectors[i][j][1];
411                 vec[2] = rVectors[i][j][2];
412
413                 //cout << vec[2] << endl;
414
415                 Ed -= mu0*Mag*Mag/(4*PI * pow(d,3)) * (3* (sx[j]*vec[0]+sy[j]*vec[1]+sz[
                   j]*vec[2])*(sx[i]*vec[0]+sy[i]*vec[1]+sz[i]*vec[2]) - (sx[j]*sx[i]+
                   sy[j]*sy[i]+sz[j]*sz[i]));
416                 //status = countdown(max.N, i, status);
417             }
418         }
419         return Ed;
420     }
421 }
422
423     double E_dipole_quick(int j)
424     {
425         double Ed = 0;
426         //int status;
427
428         for (int i = 0; i<max.N; i++)
429         {
430             if (i != j)
431             {
432                 double d = dist[i][j];
433
434                 double vec[3];
435                 vec[0] = rVectors[i][j][0];
436                 vec[1] = rVectors[i][j][1];
437                 vec[2] = rVectors[i][j][2];
438
439                 //cout << vec[2] << endl;
440
441                 Ed -= mu0*Mag*Mag/(4*PI * pow(d,3)) * (3* (sx[j]*vec[0]+sy[j]*vec[1]+sz[
                   j]*vec[2])*(sx[i]*vec[0]+sy[i]*vec[1]+sz[i]*vec[2]) - (sx[j]*sx[i]+
                   sy[j]*sy[i]+sz[j]*sz[i]));
442                 //status = countdown(max.N, i, status);
443             }
444         }
445     }
446 }

```

```

447     return Ed;
448 }
449
450
451 double E_dipole_total()
452 {
453     double Ed = 0;
454
455     omp_set_num_threads(numThreads);
456     #pragma omp parallel for reduction(+:Ed)
457     for (int i = 0; i < max_N-1; i++)
458     {
459         for (int j = i+1; j < max_N; j++)
460         {
461             Ed += dipole(i, j);
462         }
463     }
464
465     Ed /= pow((edge_N-1)*latt_const, 3);
466     return Ed;
467 }
468
469 double E_aniso_total()
470 {
471     double Ea = 0;
472
473     omp_set_num_threads(numThreads);
474     #pragma omp parallel for reduction(+:Ea)
475     for (int n = 0; n < max_N; n++)
476         //Ea += KV * (1 - pow(sx[n]*kx[n]+sy[n]*ky[n]+sz[n]*kz[n], 2));
477         Ea += KV * (0 - pow(sx[n]*kx[n]+sy[n]*ky[n]+sz[n]*kz[n], 2));
478
479     Ea /= pow((edge_N-1)*latt_const, 3);
480     return Ea;
481 }
482
483 double E_zee_total(double BBx)
484 {
485     double Ezee = 0;
486
487     omp_set_num_threads(numThreads);
488     #pragma omp parallel for reduction(-:Ezee)
489     for (int n = 0; n < max_N; n++)
490         Ezee += -Mag * BBx * sx[n];
491
492     Ezee /= pow((edge_N-1)*latt_const, 3);
493     return Ezee;
494 }
495
496
497
498 double E_surf_total()
499 {
500     double Esurf = 0;
501
502     omp_set_num_threads(numThreads);
503     #pragma omp parallel for reduction(+:Esurf)
504     for (int n = 0; n < max_N-1; n++)
505     {
506         for (int m = n+1; m < max_N; m++)
507             Esurf += sx[n]*sx[m] + sy[n]*sy[m] + sz[n]*sz[m];
508     }
509
510     Esurf *= mu0 * Mag * Mag / (2 * pow((edge_N-1)*latt_const, 6));
511
512     return Esurf;
513 }
514
515
516 double E_dipole_quick_MULT(int j)
517 {
518     double Ed = 0;
519     //int Ntrunc = max_N/num_threads;
520     //int status;
521     omp_set_num_threads(numThreads);
522     #pragma omp parallel for reduction(+:Ed)
523     for (int i = 0; i < max_N; i++)
524     {
525         Ed += dipole(i, j);
526     }
527
528     return Ed;

```

```

529 }
530
531
532
533 double E_filesave(int ed, int xx)
534 {
535     double Etotal = 0;
536
537     if (PRINT_ENERGIES)
538     {
539         string data_file_cluster = Edata_file + to_string(ed) + ".dat";
540         ofstream fout;
541         fout.open(data_file_cluster, ios::app);
542         //fout << "cgf T B M\n";
543
544         double Ed, Ea, Ezee, Esurf;
545
546         Ed = E_dipole_total();
547         Ea = E_aniso_total();
548         Ezee = E_zee_total(Bx[xx]);
549         Esurf = E_surf_total();
550         Etotal = Ed + Ea + Ezee + Esurf;
551
552         fout << xx << "\t" << T[xx] << "\t" << Bx[xx] << "\t" << Ezee << "\t" << Ea << "
553             \t" << Ed << "\t" << Esurf << "\t" << Etotal << endl;
554
555         fout.close();
556     }
557
558     return Etotal;
559 }
560
561 void detailed_measure(int ed, int xx)
562 {
563     if (DETAILED_MEASURE)
564     {
565         double m_x, m_y, m_z, MX, MY, MZ;
566         m_x = 0;
567         m_y = 0;
568         m_z = 0;
569         MX = 0;
570         MY = 0;
571         MZ = 0;
572
573         omp_set_num_threads(2);
574         #pragma omp parallel for reduction(+:m_x), reduction(+:m_y), reduction(+:m_z),
575             reduction(+:MX), reduction(+:MY), reduction(+:MZ)
576         for (int n = 0; n < max_N; n++)
577         {
578             m_x += sx[n]; // measure in x-direction
579             m_y += sy[n];
580             m_z += sz[n];
581
582             MX += abs(sx[n]); // measure total x-alignment
583             MY += abs(sy[n]);
584             MZ += abs(sz[n]);
585         }
586
587         m_x /= max_N;
588         m_y /= max_N;
589         m_z /= max_N;
590         MX /= max_N;
591         MY /= max_N;
592         MZ /= max_N;
593
594         string data_file_cluster = Mdata_file + to_string(ed) + ".dat";
595         ofstream fout;
596         fout.open(data_file_cluster, ios::app);
597         //fout << "cgf T B M\n";
598
599
600         fout << xx << "\t" << T[xx] << "\t" << Bx[xx] << "\t" << m_x << "\t" << m_y << "
601             \t" << m_z << "\t" << MX << "\t" << MY << "\t" << MZ << endl;
602
603         fout.close();
604     }
605 }
606 }
607

```

```

608
609
610 /*
611 // calculate E-dipole, but at every site i, only the local neighbours determined in local_matrix
        () are considered
612 double loc_E_dipole()
613 {
614     double Ed = 0;
615     for (int i = 0; i < max_N; i++)
616     {
617         int max = loc_POS[i][0]; // exploit that we already calculated the
            number of neighbours = number of loops
618
619         // the upper triangle matrix is already implemented in local_matrix(),
            therefore j starts always at 1 as the 0th element is #(neighbours),
            NOT an index !!
620         for (int j = 1; j < max; j++)
621         {
622             int l = loc_POS[i][j]; // l is the 'old' index when all sites
                were considered so that we can still use dist[][] and
                rVectors[][][]
623
624             // exactly the same as in E-dipole() from here.
625             double d = dist[i][l];
626
627             double vec[3];
628             vec[0] = rVectors[i][l][0];
629             vec[1] = rVectors[i][l][1];
630             vec[2] = rVectors[i][l][2];
631
632
633
634
635             Ed -= mu0*Mag*Mag/(4*PI * pow(d,3)) * (3* (sx[l]*vec[0]+sy[l]*
                vec[1]+sz[l]*vec[2])*(sx[i]*vec[0]+sy[i]*vec[1]+sz[i]*vec
                [2]) - (sx[l]*sx[i]+sy[l]*sy[i]+sz[l]*sz[i]));
            //status = countdown(max_N, i, status);
636
637         }
638     }
639     return Ed;
640 }
641 */
642
643
644
645
646
647 double measure()
648 {
649     double m_x = 0.;
650     omp_set_num_threads(numThreads);
651     #pragma omp parallel for reduction(+:m_x)
652     for (int n = 0; n < max_N; n++)
653     {
654         m_x += sx[n]; // measure in x-direction
655     }
656
657     return m_x/max_N;
658 }
659
660 array<double,2> rel_perm(double B)
661 {
662     array<double,2> res;
663     double mu_r;
664     double M;
665     double meas = measure();
666     double VOL = pow((edge_N-1)*latt_const,3);
667
668
669     //double z = mu0*max_N*measure()*Mag/(3*VOL*B);
670     M = max_N*meas*Mag/VOL;
671
672     if (B == 0)
673         mu_r = def_mur;
674     else
675     {
676         //mu_r = (1+2*z)/(1-z);
677         mu_r = 1 + mu0*M/B;
678         //mu_r = B / (B-mu0*max_N*measure()*Mag);
679
680
681     }

```

```

682
683
684     //mu_r = def_mur;
685     res[0] = mu_r;
686     res[1] = M;
687
688     return res;
689 }
690
691 double best_cutoff()
692 {
693     double best_CUTOFF = edge_N-1;
694     double curr_dist;
695
696     for (int i = 0; i < max_N-1; i++)
697     {
698         for (int j = i + 1; j < max_N; j++)
699         {
700             curr_dist = dist[i][j]/latt_const;
701
702             if (curr_dist > CUTOFF and curr_dist < best_CUTOFF)
703                 best_CUTOFF = curr_dist;
704
705             //cout << curr_dist << "\t";
706         }
707     }
708     best_CUTOFF -= 0.000001;
709     cout << "cutoff_is_" << best_CUTOFF << "_up_from_" << CUTOFF << endl;
710
711     return best_CUTOFF;
712 }
713
714
715
716 vector<int> shuffle_list()
717 {
718     vector<int> rand_list;
719     for (int i = 0; i < max_N; i++)
720         rand_list.push_back(i);
721     random_shuffle(rand_list.begin(), rand_list.end());
722     return rand_list;
723 }
724
725 void MCS_ALT(int xx, double BBx, double kt)
726 {
727     int n;
728     double E0, E1, E_a, mod_M, M_temp[3], d_M[3];
729     //int status;
730     for (n = 0; n < max_N; n++)
731     {
732         E_a = -KV * pow(sx[n]*kx[n]+sy[n]*ky[n]+sz[n]*kz[n], 2); // Anistropy energy
733         //E_d = loc_E_dipole(); // choose cutoff approximation for speed-up
734         //E_d = E_dipole(); // choose to consider ALL sites for dipole-
735                             //dipole i.a.
736         E0 = - Mag * BBx * sx[n]; // field in x-direction (Zeeman energy)
737         E0 += E_a; // sum of dipole energies
738
739         M_temp[0] = sx[n];
740         M_temp[1] = sy[n];
741         M_temp[2] = sz[n];
742
743         Marsaglia(d_M);
744
745         sx[n] += (d_m * d_M[0]);
746         sy[n] += (d_m * d_M[1]);
747         sz[n] += (d_m * d_M[2]);
748         mod_M = sqrt(pow(sx[n],2)+pow(sy[n],2)+pow(sz[n],2));
749
750         sx[n] /= mod_M;
751         sy[n] /= mod_M;
752         sz[n] /= mod_M;
753
754         E_a = -KV * pow(sx[n]*kx[n]+sy[n]*ky[n]+sz[n]*kz[n], 2); // Anistropy energy
755         //E_d = loc_E_dipole();
756         //E_d = E_dipole();
757         E1 = - Mag * BBx * sx[n]; // must be same (updated) calculation as E0
758         E1 += E_a;
759
760         if (E1 > E0)
761         {
762             if (rand0.1() > exp((E0-E1)/kt))

```

```

763             sx[n] = M_temp[0];
764             sy[n] = M_temp[1];
765             sz[n] = M_temp[2];
766         }
767     }
768
769     //status = countdown(max_N, n, status);
770
771 }
772
773 }
774
775 void MCS_one(int xx, double BBx, double kt, double mu_r, double M)
776 {
777     double E0, E1, E_a, E_d, mod_M, M_temp[3], d_M[3];
778
779     vector<int> rand_list = shuffle_list(); // experimental DO NOT USE IN THIS VERSION
780     //int status;
781     for (int m = 0; m < max_N; m++)
782     {
783         E_d = 0;
784         int n = rand_list[m]; // every superspin updated exactly once per MCS_one call,
785             but in a random fashion -> permutation
786         //int n = rnd250() % max_N;
787         //int n = m;
788
789         E_a = -KV * pow(sx[n]*kx[n]+sy[n]*ky[n]+sz[n]*kz[n], 2); // Anistropy energy
790         //E_d = loc_E_dipole(); // choose cutoff approximation for speed-up
791         if (DIPOLAR)
792         {
793             if (PERIODIC)
794             {
795                 E_d = ORF(n, mu_r, BBx, M);
796                 E0 = 0;
797                 //E_a *= (3*mu_r / (1+2*mu_r));
798             }
799             else
800             {
801                 E_d = E_dipole_quick_MULT(n);
802                 E0 = - Mag * BBx * sx[n]; // field in x-direction (Zeeman energy)
803             }
804         }
805
806         else
807             E0 = - Mag * BBx * sx[n]; // field in x-direction (Zeeman energy)
808
809
810
811         E0 += E_a; //
812         E0 += E_d; // sum of dipole energies
813
814         M_temp[0] = sx[n];
815         M_temp[1] = sy[n];
816         M_temp[2] = sz[n];
817
818         Marsaglia(d_M);
819
820         sx[n] += (d_m * d_M[0]);
821         sy[n] += (d_m * d_M[1]);
822         sz[n] += (d_m * d_M[2]);
823         mod_M = sqrt(pow(sx[n], 2) + pow(sy[n], 2) + pow(sz[n], 2));
824
825         sx[n] /= mod_M;
826         sy[n] /= mod_M;
827         sz[n] /= mod_M;
828
829         E_d = 0;
830
831         E_a = -KV * pow(sx[n]*kx[n]+sy[n]*ky[n]+sz[n]*kz[n], 2); // Anistropy energy
832         //E_d = loc_E_dipole();
833
834         if (DIPOLAR)
835         {
836             if (PERIODIC)
837             {
838                 E_d = ORF(n, mu_r, BBx, M);
839                 E1 = 0;
840                 //E_a *= (3*mu_r / (1+2*mu_r));
841             }
842             else

```

```

843         {
844             E_d = E_dipole_quick_MULT(n);
845             E1 = - Mag * BBx * sx[n]; // field in x-direction (Zeeman energy
            )
846         }
847     }
848
849     else
850         E1 = - Mag * BBx * sx[n]; // field in x-direction (Zeeman energy)
851
852
853     E1 += E_a;
854     E1 += E_d;
855
856     if (E1 > E0)
857     {
858         if (rand0_1() > exp((E0-E1)/kt))
859         {
860             sx[n] = M_temp[0];
861             sy[n] = M_temp[1];
862             sz[n] = M_temp[2];
863         }
864     }
865
866
867     //status = countdown(max_N, n, status);
868
869 }
870
871 }
872
873
874
875
876
877 void aniso_test()
878 {
879     int n;
880     double t_kx = 0.0;
881     double t_ky = 0.0;
882     double t_kz = 0.0;
883
884     double t_sx = 0.0;
885     double t_sy = 0.0;
886     double t_sz = 0.0;
887
888
889     for (n = 0; n < max_N; n++)
890     {
891         t_kx += kx[n];
892         t_ky += ky[n];
893         t_kz += kz[n];
894
895         t_sx += sx[n];
896         t_sy += sy[n];
897         t_sz += sz[n];
898     }
899
900
901     t_kx /= max_N;
902     t_ky /= max_N;
903     t_kz /= max_N;
904
905     cout << "Avg_easy_axis_components_xyz:_" << t_kx << "_" << t_ky << "_" << t_kz << endl;
906
907
908
909     t_sx /= max_N;
910     t_sy /= max_N;
911     t_sz /= max_N;
912
913     cout << "Avg_Superspin_components_xyz:_" << t_sx << "_" << t_sy << "_" << t_sz << endl;
914
915 }

```

B.1.5 perAux.h

```

1 // perAux.h
2 // additional structural functions for interacting periodic systems.
3 // intended for simulations with ORF method
4 // makes use of openMP for easily implemented multiprocessing

```

```

5 // for MC simulation
6 using namespace std;
7
8 const int perFactor = 3;
9 const int NN = 10*(pow(edge_N+2*CUTOFF,3)-pow(edge_N,3))+max_N_temp;
10
11 const double rho_bcc = 1. + pow((((double) (edge_N-1))/((double)(edge_N))), 3);
12 const double rho_fcc = 1. + 3* pow((((double) (edge_N-1))/((double)(edge_N))),2);
13
14 const int CUT_VOL = 100*pow(CUTOFF/1.5,3)+1;
15
16 double perPOS[NN][3];
17 double dist_per[max_N_temp][NN];
18 double rVectors_per[max_N_temp][NN][3];
19 int loc_POS[max_N_temp][CUT_VOL];
20
21 int max_N_per;
22 vector<int> perInd;
23
24 void sc_generator_new(int edge)
25 {
26     int n = 0;
27     double a = 1;
28
29     for (int i = 0; i < edge; i++)
30     {
31         for (int j = 0; j < edge; j++)
32         {
33             for (int k = 0; k < edge; k++)
34             {
35                 POS[n][0] = 0+a*i;
36                 POS[n][1] = 0+a*j;
37                 POS[n][2] = 0+a*k;
38
39                 n++;
40             }
41         }
42     }
43 }
44
45 void fcc_generator_new(int edge)
46 {
47     int n = 0;
48     double a = 1.;
49
50     for (int i = 0; i<edge; i++)
51     {
52         for (int j = 0; j< edge; j++)
53         {
54             for (int k = 0; k< edge; k++)
55             {
56                 POS[n][0] = 0+a*i;
57                 POS[n][1] = 0+a*j;
58                 POS[n][2] = 0+a*k;
59
60                 POS[n+1][0] = 0.5+a*i;
61                 POS[n+1][1] = 0.5+a*j;
62                 POS[n+1][2] = 0+a*k;
63
64                 POS[n+2][0] = 0.5+a*i;
65                 POS[n+2][1] = 0+a*j;
66                 POS[n+2][2] = 0.5+a*k;
67
68                 POS[n+3][0] = 0+a*i;
69                 POS[n+3][1] = 0.5+a*j;
70                 POS[n+3][2] = 0.5+a*k;
71
72                 n+=4;
73             }
74         }
75     }
76 }
77
78 void bcc_generator_new(int edge)
79 {
80     int n = 0;
81     double a = 1.;
82
83     for (int i = 0; i<edge; i++)
84     {
85         for (int j = 0; j< edge; j++)

```

```

87         {
88             for (int k = 0; k < edge; k++)
89             {
90                 POS[n][0] = 0+a*i;
91                 POS[n][1] = 0+a*j;
92                 POS[n][2] = 0+a*k;
93
94                 POS[n+1][0] = 0.5+a*i;
95                 POS[n+1][1] = 0.5+a*j;
96                 POS[n+1][2] = 0.5+a*k;
97
98                 n+=2;
99             }
100         }
101     }
102 }
103
104 void hcp_generator_new(int edge)
105 {
106     int n = 0;
107     double a = 1.;
108
109     for (int i = 0; i < edge; i++)
110     {
111         for (int j = 0; j < edge; j++)
112         {
113             for (int k = 0; k < edge; k++)
114             {
115                 POS[n][0] = (2*i+((j+k)%2))*a/2.;
116                 POS[n][1] = sqrt(3.)*(j+1./3.*(k%2))*a/2.;
117                 POS[n][2] = 2*sqrt(6.)/3.*k*a/2.;
118
119                 n+=1;
120             }
121         }
122     }
123 }
124
125 bool inn_cube(double x, double y, double z, double marg)
126 {
127     if (x < 0 + marg)
128         return false;
129     if (x > (edge_N-1)-marg)
130         return false;
131
132     if (y < 0 + marg)
133         return false;
134     if (y > (edge_N-1)-marg)
135         return false;
136
137     if (z < 0 + marg)
138         return false;
139     if (z > (edge_N-1)-marg)
140         return false;
141
142     return true;
143 }
144
145 vector<int> find_edgeInd2()
146 {
147     vector<int> EDGE;
148
149     double x,y,z;
150
151     for (int i = 0; i < max_N; i++)
152     {
153         x = POS[i][0];
154         y = POS[i][1];
155         z = POS[i][2];
156
157         if (not inn_cube(x,y,z, CUTOFF))
158             EDGE.push_back(i);
159     }
160
161     return EDGE;
162 }
163
164 bool copyable(double x, double y, double z, int cx, int cy, int cz)
165 {
166     bool trivial = (cx == 0 and cy == 0 and cz == 0);

```

```

169
170     if (inn_cube(x,y,z,-sqrt(3)*CUTOFF) and not trivial)
171         return true;
172
173     return false;
174 }
175
176
177 vector<array<double,3>> copies(array<double,3> original)
178 {
179     vector<array<double, 3>> CAND;
180     double x0 = original[0];
181     double y0 = original[1];
182     double z0 = original[2];
183
184     double x, y, z;
185
186     int sgn[3] = {-1,0,1};
187     array<double,3> cand;
188
189
190     for (int i = 0; i < 3; i++)
191     {
192         for (int j = 0; j < 3; j++)
193         {
194             for (int k = 0; k < 3; k++)
195             {
196                 x = x0 + edge_N*sgn[i];
197                 y = y0 + edge_N*sgn[j];
198                 z = z0 + edge_N*sgn[k];
199
200                 if(copyable(x,y,z, sgn[i], sgn[j], sgn[k]))
201                 {
202                     cand[0] = x;
203                     cand[1] = y;
204                     cand[2] = z;
205                     CAND.push_back(cand);
206                 }
207             }
208         }
209     }
210
211     return CAND;
212 }
213
214
215
216 double distance(int i, int j)
217 {
218     double x1, x2, y1, y2, z1, z2, r;
219
220     x1 = perPOS[i][0];
221     y1 = perPOS[i][1];
222     z1 = perPOS[i][2];
223
224     x2 = perPOS[j][0];
225     y2 = perPOS[j][1];
226     z2 = perPOS[j][2];
227
228     r = sqrt(pow(x1-x2,2)+pow(y1-y2,2)+pow(z1-z2,2));
229
230     return r;
231 }
232
233
234 void makePeriodic()
235 {
236
237     vector<int> EDGE;
238     EDGE = find_edgeInd2();
239
240
241     int len = EDGE.size();
242     int edgeInd;
243
244
245     cout << len << "_of_" << max_N << "_in_edge.\n";
246
247     if (len >= max_N - 1)
248     {
249         cout << "Superspins_will_interact_with_own_copies_-_not_physical!\n";
250         bool cont;

```

```

251         cout << "Continue_regardless?_";
252         cin >> cont;
253         if (not cont)
254             abort();
255     }
256
257     array<double, 3> orig_pos;
258     vector<array<double, 3>> copy_vector;
259
260
261     int len2;
262     int PERlen = 0;
263     double per_x, per_y, per_z;
264
265
266     for (int i = 0; i < max_N; i++)
267     {
268         perPOS[i][0] = POS[i][0];
269         perPOS[i][1] = POS[i][1];
270         perPOS[i][2] = POS[i][2];
271
272         perInd.push_back(i);
273     }
274
275
276     for (int j = 0; j < len; j++)
277     {
278         edgeInd = EDGE[j];
279
280         orig_pos[0] = POS[edgeInd][0];
281         orig_pos[1] = POS[edgeInd][1];
282         orig_pos[2] = POS[edgeInd][2];
283
284         copy_vector = copies(orig_pos);
285
286         len2 = copy_vector.size();
287         for (int i = 0; i < len2; i++)
288         {
289             per_x = copy_vector[i][0];
290             per_y = copy_vector[i][1];
291             per_z = copy_vector[i][2];
292
293
294
295             perPOS[PERlen+max_N][0] = per_x;
296             perPOS[PERlen+max_N][1] = per_y;
297             perPOS[PERlen+max_N][2] = per_z;
298
299
300             perInd.push_back(edgeInd);
301
302
303
304             PERlen++;
305         }
306     }
307
308     cout << PERlen << "_copies_determined.\n";
309
310     cout << "Index_conversion_saved.\n";
311
312     int neighb, k;
313     double d;
314     omp_set_num_threads(numThreads);
315     #pragma omp parallel for private(k, neighb, d)
316     for (int i = 0; i < max_N; i++)
317     {
318         //cout << "here\n";
319
320         neighb = 0;
321
322         for (k = 0; k < max_N + PERlen; k++)
323         {
324             d = distance(i, k);
325             if (i != k and d <= CUTOFF)
326             {
327                 neighb++;
328                 loc.POS[i][neighb] = k;
329                 dist_per[i][k] = d; // no symmetrization necessary b/c k

```

```

333                                     runs through ALL indices
334 double vec[3] = {perPOS[i][0] - perPOS[k][0], perPOS[i][1] - perPOS[k]
335                  ][1], perPOS[i][2] - perPOS[k][2]};
336 double mod = sqrt(pow(vec[0],2)+pow(vec[1],2)+pow(vec[2],2));
337 vec[0] /= mod;
338 vec[1] /= mod;
339 vec[2] /= mod;
340
341 rVectors_per[i][k][0] = vec[0];
342 rVectors_per[i][k][1] = vec[1];
343 rVectors_per[i][k][2] = vec[2];
344 }
345 }
346 loc_POS[i][0] = neighb;
347 }
348 }
349
350
351
352 cout << loc_POS[0][0] << "_neighbours_per_site." << endl;
353
354 double ORF(int i, double mu_r, double B, double M)
355 {
356
357     double Ed = 0;
358     double E_RF = 0;
359
360     double sx1 = sx[i];
361     double sy1 = sy[i];
362     double sz1 = sz[i];
363
364     double Sx = sx1;
365     double Sy = sy1;
366     double Sz = sz1;
367
368
369     int len = loc_POS[i][0];
370
371
372
373     omp_set_num_threads(numThreads2);
374     #pragma omp parallel for reduction(+:Ed), reduction(+:Sx), reduction(+:Sy), reduction(+:
375         Sz)
376     for (int j = 1; j <= len; j++)
377     {
378         int neighbInd1 = loc_POS[i][j];
379
380         int neighbInd2 = perInd[neighbInd1];
381
382         double sx2 = sx[neighbInd2];
383         double sy2 = sy[neighbInd2];
384         double sz2 = sz[neighbInd2];
385
386         Sx += sx2;
387         Sy += sy2;
388         Sz += sz2;
389
390
391         double d = dist_per[i][neighbInd1];
392
393         double vec[3];
394
395
396         vec[0] = rVectors_per[i][neighbInd1][0];
397         vec[1] = rVectors_per[i][neighbInd1][1];
398         vec[2] = rVectors_per[i][neighbInd1][2];
399
400         Ed += -mu0*Mag*Mag/(4*PI * pow(d*latt_const,3)) * (3* (sx1*vec[0]+sy1*vec[1]+sz1
401             *vec[2])*(sx2*vec[0]+sy2*vec[1]+sz2*vec[2]) - (sx1*sx2+sy1*sy2+sz1*sz2));
402     }
403
404     E_RF = -sx1*(B-0) * Mag * (3*mu_r / (1+2*mu_r));
405
406     E_RF += mu0* Mag*Mag * (sx1*Sx + sy1*Sy + sz1*Sz) * (1-mu_r)/(2*mu_r+1)/(2*PI*pow(
407         BEST_CUTOFF*latt_const,3));
408
409     if (ONSDAT)
410     {

```

```

410         ofstream ons_file;
411         ons_file.open("ons.dat", ios::app);
412         ons_file << Ed << "\t" << E_RF << endl;
413         ons_file.close();
414     }
415     return Ed + E_RF;
416 }
417 }

```

B.1.6 rnd250.c

```

1  /*
2  *  RND250.C
3  *
4  *  Zufallszahlengenerator nach dem Verfahren von Kirkpatrick und Stoll.
5  *  Dieses C-Modul enthaelt nur die globale Definition der Daten und die
6  *  Initialisierungsroutine. Der eigentliche Zufallszahlengenerator ist
7  *  als Makro in der Datei RND250.H kodiert.
8  *
9  *  Implementation: Ralf Meyer, Fred Hucht
10 *  Version          : 2.0
11 *  entwickelt am   : 14. Februar 1995
12 *
13 *  Copyright (c) 1995 Ralf Meyer, 47058 Duisburg, Germany
14 */
15
16 #include "rnd.h"
17 struct st_rnd250 Rnd250;
18
19
20 void
21 seed250 (long seed)
22 /*
23 *  Initialisiert den Zufallszahlengenerator. Hierzu wird ein Modulo-
24 *  Zufallszahlengenerator benutzt. Um die lineare Unabhaengigkeit der
25 *  einzelnen Bits zu garantieren werden nachtraeglich noch Bitmasken
26 *  einem Teil der Daten ueberlagert. Um die dadurch verursachten An-
27 *  fangskorrelationen zu beseitigen werden die ersten Zufallszahlen
28 *  verworfen.
29 *
30 *  Parameter:
31 *  seed — Startwert
32 */
33 {
34     int i;
35     long j, k;
36
37     if (seed < 1) /* keine negativen Startwerte */
38         seed = 1;
39
40     for (i=0; i<250; ++i) { /* Schieberegister mit Zufallszahlen fuellen */
41         k = seed / 127773;
42         seed = 16807 * (seed - k*127773) - 2836*k;
43         if (seed < 0)
44             seed += 0x7FFFFFFF;
45         Rnd250.field[i] = seed;
46     }
47
48     k = 0x7FFFFFFF; /* Masken ueberlagern */
49     j = 0x40000000;
50     for (i=1; i<250; i+=8)
51         Rnd250.field[i] = (Rnd250.field[i] & k) | j;
52
53     Rnd250.point = 249; /* Zeiger initialisieren */
54
55     for (i=0; i<4711; ++i) /* Anfangszahlen verworfen */
56         rnd250();
57 }

```

B.1.7 random-spd5.h

```

1  //
2  // random-spd5.h
3  //
4  // functions that require random number generators are collected here
5  // intended to use rnd.h by Ralf Meyer
6
7  const double PI = 3.1415926535897932385;
8

```

```

9  double GaussDis(double, double, double);
10
11
12  const double real_RND250_MAX = (double) MAXRAND250;
13
14  /*
15      generate a random number in [0,1)
16  */
17  double rand0_0999()
18  {
19      return (rnd250() / (real_RND250_MAX+1.0));
20  }
21
22
23  /*
24      generate a random number in (0,1)
25  */
26  double rand0001_0999()
27  {
28      return ((rnd250()+1.0) / (real_RND250_MAX+2.0));
29  }
30
31
32  /*
33      generate a random number in [0,1]
34  */
35  double rand0_1()
36  {
37      return (rnd250() / real_RND250_MAX);
38  }
39
40
41
42  /*
43      yield a 3D random unit vector
44  */
45  void Marsaglia(double *V)
46  {
47      double rsq, y1, y2;
48      do
49      {
50          y1 = rand0001_0999() * 2.0 - 1.0;
51          y2 = rand0001_0999() * 2.0 - 1.0;
52          rsq = y1 * y1 + y2 * y2;
53      }
54      while (rsq > 1.0);
55      V[0] = 2.0 * y1 * sqrt((1.0 - rsq));
56      V[1] = 2.0 * y2 * sqrt((1.0 - rsq));
57      V[2] = 1.0 - 2.0 * rsq;
58  }
59
60  double GaussDis(double x, double avg, double sig)
61  {
62      double fct;
63
64      fct = (1.0/(sqrt(2.0*PI)*sig))*exp(-0.5*(x-avg)*(x-avg)/(sig*sig));
65
66      return(fct);
67  }
68
69
70  /*
71      yields a 3D unit vector, but with Gauss-Distribution in y,z-coordinates
72      <-> anisotropy axis distribution - easy axis in x- direction
73
74
75  void AnisotropyAxis(double *V)
76  {
77      double rsq, y, z, y2, z2, ss;
78      do
79      {
80          do
81          {
82              y = rand0_1() * 2.0 - 1.0;
83              y2 = rand0_1();
84          }
85          while (y2 > GaussDis(y, y_avg, y_sig));
86          do
87          {
88              z = rand0_1() * 2.0 - 1.0;
89              z2 = rand0_1();
90          }

```

```

91         while (z2 > GaussDis(z, z_avg, z_sig));
92         rsq = y*y + z*z;
93     }
94     while (rsq > 1.0);
95     ss = rand0_1() * 2.0 - 1.0;
96     V[0] = (ss/fabs(ss))*sqrt(1.0-rsq);
97     V[1] = y;
98     V[2] = z;
99 }
100 */

```

B.2 domainFinder.cpp

This is the programme used to generate the visualizations of 3D spin landscapes that are presented in the later parts of the thesis. This programme finds sublattices in a data set of spin positions and -orientations which are parallel, and analyzes size and orientation of all theses sublattices within one set. In conjunction with *gnuplot*, an entire set of spins can be visualized and via colorization by the programme, these images can be interpreted by the user even if thousands of spins in three dimensions are displayed at once.

Algorithm design and implementation in C++11 were done by the author.

```

1  #include <iostream>
2  #include <fstream>
3  #include <sstream>
4  #include <cmath>
5  #include <vector>
6  #include <array>
7  #include <string>
8  #include <algorithm>
9  #include <thread>
10 #include <chrono>
11
12 #include <stdlib.h>
13 // #include <omp.h>
14
15 using namespace std;
16
17 string data_file = "spin1000.dat"; // Input file name
18 string ofile_name = "showDomain.dat"; // gnuplot plotable coordinates of all vectors in a LARGE
    domain
19 string ofile2_name = "DomNegative.dat"; // same as above, only reduced to "interesting" domain
    PAIRS
20 string gnu_file = "colorDom.gnu"; // file name of gnu-script
21
22 const int max_N = 4*8*8*8; // Number of lines registered in input file == number of vectors
    considered
23
24 double latt_const = 1;
25 double POS[max_N][3]; // position vectors of sites
26 double VEC[max_N][3]; // orientation vectors of sites
27 bool forbidden[max_N]; // flag if a site has yet to be assigned to a domain
28 bool neg[max_N]; // flag if a site should be printed in negative file
29 double dist[max_N][max_N]; // distances btw sites
30 double MAX_DIST = 10.; // maximum distance btw to sites that are considered neighbours
31 //double MIN_CORR = -1.;
32 double MIN_CORR = cos(20*3.142/180.); // minimum scalar product btw to orientations that are
    considered belonging to same domain
33 double ACC;
34 double min_size = 80; // minimum size of a 'domain' that is significant enough to be stored in
    LARGEDOMAINS
35 //double perp_corr = 2.;
36 double perp_corr = cos(85*3.142/180.); // minimum scalar product btw to domains that are
    considered antiferromagnetic pairs
37 int dcounter; // stores number of domains that have been identified
38 int mem_counter; // stores number of member in one such domain
39 vector<vector<int>> DOMAINS; // stores every index vector of all small (!!) domains
40
41
42 //read data files and fill position and vector arrays
43 void read_data(string filename, int num)
44 {
45     ifstream data;

```

```

46     data.open(filename);
47     std::string line;
48
49     for (int i = 0; i < num*max.N; i++)
50     {
51
52         getline(data, line);
53         double col1, col2, col3, col4, col5, col6;
54
55         istringstream ss(line);
56         ss >> col1 >> col2 >> col3 >> col4 >> col5 >> col6;
57     }
58
59     int counter = 0;
60     for (int j = num*max.N; j < (num+1)*max.N; j++)
61     {
62
63         getline(data, line);
64         double col1, col2, col3, col4, col5, col6;
65
66         istringstream ss(line);
67         ss >> col1 >> col2 >> col3 >> col4 >> col5 >> col6;
68
69         POS[counter][0] = col1;
70         POS[counter][1] = col2;
71         POS[counter][2] = col3;
72
73         VEC[counter][0] = col4;
74         VEC[counter][1] = col5;
75         VEC[counter][2] = col6;
76         counter++;
77     }
78
79     data.close();
80 }
81
82
83
84 // calculate distance btw two positions
85 double distance(int i, int j)
86 {
87     double x1, x2, y1, y2, z1, z2;
88
89     x1 = POS[i][0];
90     y1 = POS[i][1];
91     z1 = POS[i][2];
92
93     x2 = POS[j][0];
94     y2 = POS[j][1];
95     z2 = POS[j][2];
96
97     return sqrt(pow(x1-x2,2) + pow(y1-y2,2) + pow(z1-z2,2));
98 }
99
100 // scalar product btw two vectors
101 double scp(int i, int j)
102 {
103     double vx1, vx2, vy1, vy2, vz1, vz2;
104
105     vx1 = VEC[i][0];
106     vy1 = VEC[i][1];
107     vz1 = VEC[i][2];
108
109     vx2 = VEC[j][0];
110     vy2 = VEC[j][1];
111     vz2 = VEC[j][2];
112
113     return vx1*vx2+vy1*vy2+vz1*vz2;
114 }
115
116 // calculate center of mass of a given vector of sites
117 vector<double> POS_center(vector<int> sample)
118 {
119     double center[3];
120     vector<double> result;
121     center[0] = 0;
122     center[1] = 0;
123     center[2] = 0;
124
125     int len = sample.size();
126
127     for (int i = 0; i < len; i++)

```

```

128     {
129         center[0]+=POS[sample[i]][0];
130         center[1]+=POS[sample[i]][1];
131         center[2]+=POS[sample[i]][2];
132     }
133
134     result.push_back(center[0]/len);
135     result.push_back(center[1]/len);
136     result.push_back(center[2]/len);
137
138     return result;
139 }
140
141 // calculate distance of two vectors (NOT indices)
142 double center_dist(vector<double> a, vector<double> b)
143 {
144     double x1, x2, y1, y2, z1, z2;
145
146     x1 = a[0];
147     y1 = a[1];
148     z1 = a[2];
149
150     x2 = b[0];
151     y2 = b[1];
152     z2 = b[2];
153
154     return sqrt(pow(x1-x2,2) + pow(y1-y2,2) + pow(z1-z2,2));
155 }
156
157 // ignore vectors that aren't unit vectors.
158 void test_data()
159 {
160     for (int i = 0; i < max.N; i++)
161         if (scp(i,i) < 0.99 or scp(i,i) > 1.01)
162         {
163             VEC[i][0] = 0;
164             VEC[i][1] = 0;
165             VEC[i][2] = 0;
166
167             cout << i+1 << "_ignored!\n";
168         }
169 }
170
171 // fill distance matrix
172 void dist_matrix()
173 {
174     //upper triangle matrix
175     for (int i = 0; i < max.N-1; i++)
176     {
177         for (int j = i+1; j < max.N; j++)
178             dist[i][j] = distance(i,j);
179     }
180
181     // symmetry i<->j
182     for (int i = 0; i < max.N-1; i++)
183     {
184         for (int j = i+1; j < max.N; j++)
185             dist[j][i] = dist[i][j];
186     }
187 }
188
189 // return list of neighbours within max_dist radius
190 vector<int> neighbours(int i, double max_dist)
191 {
192     vector<int> neighb;
193     for (int j = 0; j < max.N; j++)
194     {
195         if (dist[i][j] <= max_dist)
196         {
197             //cout << i << " NB von " << j << endl;
198             neighb.push_back(j);
199         }
200     }
201     return neighb;
202 }
203
204 // Calculate avg direction of a domain given its index in DOMAINS
205 vector<double> vec_mean(int domain)
206 {

```

```

210     double new_mean[3];
211     new_mean[0] = 0;
212     new_mean[1] = 0;
213     new_mean[2] = 0;
214
215     vector<double> result;
216
217     int len = DOMAINS[domain].size();
218
219     for (int i = 0; i < len; i++)
220     {
221         new_mean[0] += VEC[DOMAINS[domain][i]][0];
222         new_mean[1] += VEC[DOMAINS[domain][i]][1];
223         new_mean[2] += VEC[DOMAINS[domain][i]][2];
224     }
225     /*
226     new_mean[0] = pow(new_mean[0], 1./len);
227     new_mean[1] = pow(new_mean[1], 1./len);
228     new_mean[2] = pow(new_mean[2], 1./len);
229     */
230     double mod = sqrt(pow(new_mean[0], 2) + pow(new_mean[1], 2) + pow(new_mean[2], 2));
231     for (int i = 0; i < 3; i++)
232         result.push_back(new_mean[i]/mod);
233
234     return result;
235
236 }
237
238 // scalar product of a given vector old and a vector at index new_ind
239 double scp_vec_ind(vector<double> old, int new_ind)
240 {
241     double vx1, vx2, vy1, vy2, vz1, vz2;
242
243     vx1 = old[0];
244     vy1 = old[1];
245     vz1 = old[2];
246
247     vx2 = VEC[new_ind][0];
248     vy2 = VEC[new_ind][1];
249     vz2 = VEC[new_ind][2];
250
251     return vx1*vx2+vy1*vy2+vz1*vz2;
252 }
253
254 // scalar product of two directly given vectors
255 double dom_dom(vector<double> a, vector<double> b)
256 {
257     double vx1, vx2, vy1, vy2, vz1, vz2;
258
259     vx1 = a[0];
260     vy1 = a[1];
261     vz1 = a[2];
262
263     vx2 = b[0];
264     vy2 = b[1];
265     vz2 = b[2];
266
267     return vx1*vx2+vy1*vy2+vz1*vz2;
268 }
269
270
271 // return list of "good" neighbours among neighbours
272 vector<int> good_neighbours(vector<double> dom_mean, vector<int> neighb, double min_corr, int
    domain)
273 {
274     int n;
275
276     int len = neighb.size();
277     vector<int> goodNs;
278     for (int i=0; i<len; i++)
279     {
280         n = neighb[i];
281         //if (scp(n, j) >= min_corr and not forbidden[n])
282         if (scp_vec_ind(dom_mean, n) >= min_corr and not forbidden[n])
283         {
284
285             forbidden[n] = true;
286             DOMAINS[domain].push_back(n);
287             //cout << n << " found as good neighbor\n";
288             goodNs.push_back(n);
289
290         }

```

```

291     }
292     // flag if a site is isolated = w/o a single good neighbour = break recursive loop in
        findDomain()
293     if (goodNs.size() == 0)
294         goodNs.push_back(-1);
295
296     return goodNs;
297 }
298
299
300 // Central algorithm: Recursively, find 'good neighbours' among connected and not yet domain-
        associated vectors
301 void findDomain(int start, double max_dist, double min_corr, int domain)
302 {
303     vector<int> goodNs;
304     vector<int> neighb;
305     vector<double> dom_mean;
306
307     int gn_size;
308     int new_start;
309
310     // continuous update of what would be considered a good neighbour from avg (!!!)
        orientation of current domain members
311     if (DOMAINS[domain].size() >= 1)
312         dom_mean = vec_mean(domain);
313     else
314     {
315         dom_mean.push_back(VEC[start][0]);
316         dom_mean.push_back(VEC[start][1]);
317         dom_mean.push_back(VEC[start][2]);
318     }
319
320     neighb = neighbours(start, max_dist);
321     goodNs = good_neighbours(dom_mean, neighb, min_corr, domain);
322     gn_size = goodNs.size();
323
324     for (int i = 0; i < gn_size; i++)
325     {
326         new_start = goodNs[i];
327         if (new_start != -1)
328         {
329             //cout << "start findDomain from " << new_start << endl;
330
331             findDomain(new_start, max_dist, min_corr, domain);
332         }
333     }
334 }
335
336
337 // print a single vector's coordinates for gnuplot to ofile_name (!!)
338 void print_coord(int i)
339 {
340     ofstream ofile;
341     ofile.open(ofile_name, ios::app);
342
343     double x, y, z, vx, vy, vz;
344     double scale = 0.5;
345
346     vx = scale * VEC[i][0];
347     vy = scale * VEC[i][1];
348     vz = scale * VEC[i][2];
349
350     x = POS[i][0] - 0.5*vx;
351     y = POS[i][1] - 0.5*vy;
352     z = POS[i][2] - 0.5*vz;
353
354
355     ofile << x << "\t" << y << "\t" << z << "\t" << vx << "\t" << vy << "\t" << vz << endl;
356
357     ofile.close();
358
359 }
360
361 // print a single vector's coordinates for gnuplot to ofile2_name (!!)
362 void print_coord_alt(int i)
363 {
364     ofstream ofile2;
365     ofile2.open(ofile2_name, ios::app);
366
367     double x, y, z, vx, vy, vz;
368
369     vx = VEC[i][0];

```

```

370     vy = VEC[i][1];
371     vz = VEC[i][2];
372
373     x = POS[i][0] - 0.5*vx;
374     y = POS[i][1] - 0.5*vy;
375     z = POS[i][2] - 0.5*vz;
376
377
378     ofile2 << x << "\t" << y << "\t" << z << "\t" << vx << "\t" << vy << "\t" << vz << endl;
379
380     ofile2.close();
381
382 }
383
384 // generate the complete gnuplot script to visualize all large domains
385 void gnu_script(vector<vector<int>>> data)
386 {
387     ofstream gnu;
388     gnu.open(gnu_file);
389
390     vector<int> param;
391
392     int lines = data.size();
393
394     for (int i = 0; i < lines; i++)
395         param.push_back(data[i].size());
396
397
398     gnu << "reset" << endl;
399     gnu << "set_view_equal_xyz" << endl;
400     gnu << "set_title_" << endl;
401     gnu << "splot_";
402
403     char c = ' ';
404
405
406     int von = 0;
407     int bis = param[0]-1;
408
409     for (int i = 0; i < lines; i++)
410     {
411
412         gnu << " " << ofile_name << " 'Lu_1:2:3:4:5:6_every_1::";
413         gnu << von << " :;" << bis << "_with_vectors_";
414         gnu << "title_" << c << "Sublattice_" << i+1 << c << ",_";
415
416         von += param[i];
417         bis += param[i+1];
418     }
419
420     gnu << endl;
421
422     gnu.close();
423 }
424
425
426
427 // MAIN FUNCTION
428 int main()
429 {
430
431     int number;
432
433     cout << "Which_part?_";
434     cin >> number;
435     // Some initializations.
436     dcounter=0;
437
438     ofstream ofile;
439     ofstream ofile2;
440     ofstream gnu;
441     ofile.open(ofile_name, ios::trunc);
442     ofile.close();
443     ofile2.open(ofile2_name, ios::trunc);
444     ofile2.close();
445     gnu.open(gnu_file, ios::trunc);
446     gnu.close();
447
448     cout << "Output_files_reset!\n";
449
450     vector<vector<int>>> LARGEDOMAINS;
451     vector<vector<double>>> LARGEDOMAINS_AXIS;

```

```

452
453 // read and test input file
454
455
456 read_data(data_file , number);
457 test_data();
458 cout << "data_file_read...\n";
459
460
461 // Every spin is eligible for a domain and all distances are stored
462 for (int i = 0; i < max.N; i++)
463 {
464     neg[i] = true;
465     forbidden[i] = false;
466 }
467 dist_matrix();
468 cout << "distances_calculated...\n";
469
470 // Necessary initialization of vectors
471 vector<int> placeholder;
472 DOMAINS.push_back(placeholder);
473
474 //int domainSize;
475
476 // Call the central algorithm until every site has been assigned to an element of
477 // DOMAINS or has been found as isolated
478 for (int i = 0; i < max.N; i++)
479 {
480     //cout << "Test " << i << endl;
481     if (not forbidden[i])
482     {
483         DOMAINS.push_back(placeholder);
484         dcounter++;
485         findDomain(i, MAX_DIST, MIN_CORR, dcounter);
486     }
487 }
488
489 cout << "How_accurate?_";
490 cin >> ACC;
491
492 // From the partitioning, remove deviating spins and find LARGE domains (minimum number
493 // of members) and store them together with their orientation in LARGEDOMAINS
494
495 double ratio = 0;
496 int DCOUNTER = 0;
497
498 double x, y, z;
499 vector<double> dom_mean;
500 int DOMsize = DOMAINS.size();
501
502 for (int i = 0; i < DOMsize; i++)
503 {
504     vector<int> v_candidate = DOMAINS[i];
505     int len_candidate = v_candidate.size();
506     dom_mean = vec_mean(i);
507
508     vector<int> v;
509
510     // remove/don't add spins that deviate too much from mean direction
511     for (int j = 0; j < len_candidate; j++)
512     {
513         if (scp_vec_ind(dom_mean, v_candidate[j]) >= ACC * sqrt((1+MIN_CORR)/2))
514         {
515             v.push_back(v_candidate[j]);
516             neg[v_candidate[j]] = false;
517         }
518     }
519
520     int len = v.size();
521
522     // find best sets with significant size
523     if (len >= min_size)
524     {
525         LARGEDOMAINS.push_back(v);
526         DCOUNTER++;
527         ratio += len;
528
529         LARGEDOMAINS_AXIS.push_back(dom_mean);
530         x = dom_mean[0];
531         y = dom_mean[1];
532         z = dom_mean[2];
533     }
534 }

```

```

532
533
534         cout << "Sublattice_" << DOUNTER << "_of_size_" << len << "_w/_
            direction_" << x << "_" << y << "_" << z << endl;
535
536         for (int j = 0; j < len; j++)
537             print_coord(v[j]);
538         //ofile << endl;
539     }
540
541 }
542
543
544
545
546
547     ratio /= max_N;
548
549     cout << DOUNTER << "_sublattices_found!\n";
550     cout << "Sublattice_in/out_ratio_=" << ratio << endl;
551
552
553     // Among the significant domains, find "interesting" domains, i.e. w/ anti-parallel or
        perpendicular orientation
554
555
556     int large_size = LARGEDOMAINS.size();
557
558
559     int pair_counter = 0; // count all pairs (perp, parall, anti-parall)
560     int anti_counter = 0; // count anti-pairs
561     int red_counter; // count almost parallel pairs
562     int RED_COUNTER = 0;
563     if (large_size >= 2)
564     {
565         for (int i = 0; i < large_size-1; i++)
566         {
567             red_counter = 0;
568
569             for (int j = i+1; j < large_size; j++)
570             {
571                 vector<double> a = LARGEDOMAINS_AXIS[i];
572                 vector<double> b = LARGEDOMAINS_AXIS[j];
573                 vector<int> v1 = LARGEDOMAINS[i];
574                 vector<int> v2 = LARGEDOMAINS[j];
575                 //if (center_dist(POS_center(v1), POS_center(v2)) <= min_size*
                    MAX_DIST)
576                 if (dom_dom(a,b) <= - ACC * sqrt(0.5*(MIN_CORR+1)) or abs(
                    dom_dom(a,b)) <= perp_corr)
577                 {
578                     pair_counter++;
579                     cout << i+1 << "_vs_" << j+1 << "_interesting!";
580                     if (dom_dom(a,b) <= - ACC * sqrt(0.5*(MIN_CORR+1)))
581                     {
582                         cout << "_(anti-parr)\n";
583                         anti_counter++;
584                     }
585                     else
586                         cout << "_(perpendicular)\n";
587
588                     /*
589                     int len1 = v1.size();
590                     int len2 = v2.size();
591                     for (int k = 0; k < len1; k++)
592                         print_coord_interesting(v1[k]);
593                     for (int k = 0; k < len2; k++)
594                         print_coord_interesting(v2[k]);
595                     */
596                 }
597             }
598             else
599             {
600                 if (dom_dom(a,b) >= + ACC * sqrt(0.5*(MIN_CORR+1)))
601                 {
602                     pair_counter++;
603                     cout << i+1 << "_vs_" << j+1 << "_interesting!";
604                     cout << "_(almost_parr)\n";
605                     red_counter++;
606                 }
607             }
608         }
609     }

```

```

610             RED.COUNTER += red_counter;
611
612
613         }
614     }
615
616     if (2* anti_counter == large_size - RED.COUNTER)
617         cout << "!!!_Each_sublattice_has_exact_antiparallel_partner_!!!\n";
618 }
619
620
621
622 // In case any large domains were found, produce the complete gnuscript for colored
623 // visualization
624 cout << pair_counter << "_interesting_pairs.\n";
625
626 if (large_size >= 1)
627 {
628     gnu_script(LARGEDOMAINS);
629
630     for (int i = 0; i < max_N; i++)
631     {
632         if (neg[i])
633             print_coord_alt(i);
634     }
635 }
636
637 return 0;
638
639 }
```

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Ich versichere, dass ich die Arbeit selbstständig verfasst und keine anderen als die angegebenen Quellen und Hilfsmittel benutzt sowie Zitate kenntlich gemacht habe.

Aachen, den